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Direct Air Capture
and Negative Emission Technologies
in Deep Mitigation Pathways

in collaboration with

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Non è strada di chi parte e già vuole arrivare,
Non la strada dei sicuri, dei sicuri di riuscire
Non è fatta per chi è fermo, per chi non vuol cambiare
E' la strada di chi parte ed arriva per partire.

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Giulia

Abstract

Large scale deployment of Negative Emission Technologies (NETs) have been shown to be key to attain stringent climate stabilization targets, as shown in the IPCC AR5 and recent analysis of 1.5°C-consistent pathways. Much criticism has been directed at the sustainability and feasibility of large-scale Bio-Energy with Carbon Capture and Storage (BECCS) deployment due to interaction with food and water security. Therefore, additional NET options, such as Direct Air Capture (DAC), should be investigated in a combined portfolio analysis. DAC can offset decentralized emissions, thus reducing the mitigation effort in energy-intensive sectors. In this work, I explore the role of different technological options for DAC using TIAM, a global Integrated Assessment Model (IAM), with different carbon budget constraints.

The contribution of this work is (1) to investigate the current state of the art for Direct Air Capture Technologies, combining technical designs found in the literature with first pilot plants and the perspective of companies behind them. (2) Develop an Expert Elicitation questionnaire to understand the most promising technologies to be investigated further, given the fragmentary literature available. (3) Implement DAC processes in TIAM, accounting for possible interactions with other elements of the energy sector, so (4) to investigate its role as part of an integrated NET portfolio, including also BECCS and afforestation, in deep decarbonization pathways consistent with 2°C and 1.5°C temperature increase. (5) Extensive sensitivity was performed to check results robustness with respect to a number of parameters, given the high uncertainties related to them. Results suggest that DAC will allow to reduce the mitigation effort and the related costs in 1.5 and 2°C scenarios, requiring less drastic decarbonization in the mid term. Although DAC itself would be deployed at mass scale only in the second half of the century, its large potential for capturing CO2 emissions is such that it impacts the short term decarbonization strategies, allowing fossil fuel to play a role in the electricity mix up to 2050 and containing the share of intermittent renewable generation. Moreover, DAC is shown to complement other types of NET, such as biomass with CCS, rather than compete with them. Despite its significant potential, actual DAC deployment is subject on a variety of factors, especially the rate at which capacity will be able to installed. Other factors, such as energy use and capital costs, appear to be less relevant. Further work to estimate technical and social constraints is thus deemed.

Keywords: Direct Air Capture, Negative Emission Technologies, Climate Change Mitigation, Integrated Assessment Models, Expert Elicitation

Sommario

Ambiziosi scenari di mitigazione per mantenere l'aumento di temperatura al di sotto di 2°C o 1.5°C richiedono lo sviluppo su larga scala di tecnologie per realizzare livelli negativi di emissioni (NETs). Molte critiche vengono rivolte alla sostenibilità e fattibilità di uno sviluppo massivo di BECCS (i.e. impianti a biomassa con successiva cattura di CO_2), visto l'estensivo utilizzo di risorse quali acqua e terreno. All'interno di una strategia diversificata, tecnologie per catturare CO_2 dall'atmosfera (DAC - Direct Air Capture) ricopriranno perciò un ruolo strategico, con il vantaggio di abbattere anche le emissioni decentralizzate, in settori difficili e costosi da decarbonizzare altrimenti, come il trasporto e l'industria intensiva. Questo lavoro di tesi vuole valutare il ruolo di diverse tecnologie per DAC utilizzando TIAM, un modello integrato per rappresentare il sistema energetico e le dinamiche economiche a livello globale.

Il contributo di questa ricerca è (1) analizzare lo stato dell'arte su DAC, utilizzando le informazioni disponibili in letteratura e i dati provenienti dai primi impianti pilota, e (2) combinarle con opinioni di esperti nel settore, attraverso lo sviluppo di un Expert Elicitation, in modo da identificare le soluzioni tecnologiche più promettenti, vista la frammentarietà delle fonti. (3) Implementare in TIAM diverse opzioni per realizzare DAC, considerando possibili interazioni con il resto del sistema energetico e (4) analizzare il loro ruolo insieme ad altre strategie di mitigazione, in particolare BECCS e forestazione, in scenari consistenti con 2°C e 1.5°C, per poi (5) sviluppare una dettagliata analisi di sensitività e valutare l'impatto di una serie di variabili tecnico-economiche, vista l'incertella legata a queste. I risultati suggeriscono che DAC permetterà di ridurre gli sforzi di mitigazione e i relativi costi in scenari di 1.5 e 2°C, richiedendo una decarbonizzazione meno drastico nel medio periodo. Anche se DAC verrà sviluppata su larga scala solo nella seconda metà del secolo, la possibilità di catturare grandi quantità di CO_2 è tale da avere notevoli impatti sulle strategie di mitigazione messe in atto nel breve periodo, permettendo di ritardare il *phase-out* completo dai combustibili fossili per la generazione elettrica dopo il 2050, e riducendo la quantità di generazione rinnovabile intermittente. Inoltre, lo sviluppo di DAC appare complementare a quello di altre NET, tra cui BECCS, invece che competere con loro. Nonostante il notevole potenziale, lo sviluppo futuro di DAC è influenzato da una serie di fattori, soprattutto il tasso di crescita della capacità installata, mentre il fabbisogno energetico e i costi appaiono meno rilevanti , richiedendo ulteriori studi.

Parole Chiave: Direct Air Capture, Negative Emission Technologies, Mitigazione dei Cambiamenti Climatici, Integrated Assessment Models, Expert Elicitation

Executive Summary

General Framework

The Paris Climate Agreement has set ambitious climate targets such as keeping global warming well below 2°C with respect to pre-industrial levels. These objectives require extremely deep and fast decarbonization of the power sector, as well as drastic emission reduction in all other energy intensive sectors.

Keeping temperature below the set target ultimately requires becoming carbon neutral and keeping carbon budgets in check [1]: considering the current level of emissions close to 40 Gt/yr and the delay of global mitigation efforts, the scientific community has recently focused on the potential for large scale GHG removal from the atmosphere via Negative Emission Technologies (NETs) as a way to reduce the carbon stock inherited [2]. NETs can achieve a global net removal of CO2 from the atmosphere, thus offsetting emissions that were released in the past or in the future; moreover, they can compensate for current emissions from sources which are difficult to mitigate directly, such as the transportation, industry or agricultural sector. Without NET, a rapid and massive expansion of intermittent renewable sources is needed to meet Paris agreement target in 2050 and further keep temperature increase below 2°C by end of century, with an impact on grid security, storage and additional cost for the entire system. Studies on NETs, also referred to as Carbon Dioxide Removal (CDR), has been conducted for almost two decades, but the topic has received more attention since the IPCC's Fifth Assessment Report (AR5) was published in 2013 [2], and will gain even more relevance as research is focusing on stringent mitigation targets, limiting the temperature increase to 1.5°C, as it will be investigated in the new IPCC report issued next November.

State of the Art

Bioenergy with Carbon Capture and Storage (BEECS) and afforestation are typically considered the most attractive options to realize negative carbon emissions, given the amount of carbon stored in biomass. However, concerns about the sustainability of bioenergy and side effects on food security, water use, and ecosystems, have led many experts to focus on other ways to capture CO_2 from the atmosphere. Direct Air Capture (DAC) is a complementary technology, since it can capture the CO_2 produced by distributed sources such as residential heating/cooling and transportation, which appear difficult and expensive to be decarbonized. In addition, this technology will reduce the impact in term of water and land use compared to biological CDR strategies and the sustainability implications of large-scale bioenergy deployment [3].

Many studies that have evaluated the amount of negative emissions achievable through the deployment of BECCS or DAC, considered them in individually [4, 5, 6], not focusing on their mutual interaction and the role of these technologies within the broader energy system.

This research work aims to couple a techno-economic analysis of the design of DAC plants, with an evaluation of its role in future mitigation scenarios through the use of Integrated Assessment Models (IAMs). Extensive sensitivity analyses has been conducted with respect to a number of the key parameters in order to ensure the robustness of our results, such as energy consumption, cost estimates, diffusion rate and competition with other NETs. The main goal, therefore, is not to provide specific levels of the policy variables, but rather to assess the optimal mitigation portfolio and identify the crucial parameters affecting it. By understanding the effect of different parameters on the deployment of DAC technologies, the final aim is to help governments and firms decide among different investment strategies for CO_2 capture technologies.

Methodology

Integrated Assessment Models (IAMs), or energy system models, are the main methodological tools used in this work. Within the energy and climate policy science, IAMs have been developed to represent the complex interactions between climate science, economics and the energy systems, so to assess the feasibility of a range of mitigation pathways and inform policy-maker about the urgency of policy implementation and government support. For this research work, the Times Integrated Assessment Model (TIAM) - maintained by the Grantham Institute at Imperial College London - has been used. A number of new processes and commodities have been implemented in TIAM framework so to represent Direct Air Capture plants and their interaction with the wider energy system and with other NET strategies, such as BECCS and afforestation. Three different technologies has been added to TIAM model to represent DAC plants: in order to characterize them in terms of cost parameters and energy inputs, an additional commodity has been created to model the possibility use of waste heat to fuel these processes, coming from recovery from industrial activities (iron and steel, cement, pulp and paper sectors) and from low-carbon power plants (CHP from Concentrated Solar Power and nuclear plants). Moreover, afforestation has been model as an additional process to achieve negative emissions and techno-economic parameters have been updated for BECCS plants according to recent reviews. The stream of CO2 captured by DAC plants has been modeled with a separate commodity with respect to other sequestration options (BECCS, traditional CCS in electricity and industrial sector) so to track this technology individually and investigate the impact of different transport cost to represent the location flexibility offered by DAC, as it does not need to be coupled with point emissions sources. Impact assessment of DAC in terms of land, water and material use has been performed ex-post.
At the same time, given the uncertainties related to this technology in term of energy and cost parameter, and future possible upscaling, an extensive robustness analysis has

been conducted on a number of key variables.

As additional tool, an Expert Elicitation has been carried out to better understand the potential of this new technology in the future from the perspective of specialists currently researching in this field, given the fragmentary and limited literature available on this topic.

Structure

This thesis work is organized as follows. I begin with a general introduction on the motivations behind this study within the climate change framework (Chapter 1), and then discuss the current state of the art for DAC options (Chapter 2), combining techno-economic assessments found in the literature on energy and cost parameters with first demonstration plants and commercial companies. Then, I describe the methodology behind this study (Chapter 3), presenting IAMs and the model TIAM used in this work, while discussing the development of an Expert Elicitation questionnaire. Later on, in Chapter 4 the implementation of DAC technologies within the TIAM modeling framework is discussed. Finally, the results obtained from a wide range of scenarios is discussed: the role of DAC as part of a NET portfolio both in 2°C and 1.5°C mitigation pathways (Chapter 5), followed by extensive sensitivity analyses (Chapter 6), including results and discussion of the impact of different parameters on the deployment of DAC and other mitigation strategies throughout the century. Chapter 7 summarizes the main findings of the research work, including suggestions for future development.

Main Results and Lesson Learnt

DAC is still in its early stage so that no technological convergence has been reached yet: different plant designs and sorbent materials are being tested both at lab scale and in first demonstration plants, trying to identify the most suitable one.

Considering also experts opinion through the expert elicitation, the designs which are closer to the commercial scale are the one based on hydroxide solutions (named DAC1), currently investigated by Carbon Engineering company, and the one adopting amine-modified solid adsorbents (referred to as DAC2), as it is being developed by Climeworks in Switzerland and Global Thermostat in California. All these companies are currently running their pilot plants and they claim to be able to bring DAC plants on the market by 2025, making it a competitive technology also from a commercial perspective, beside its mitigation role. Their cost estimates are still quite high, around 200-300 \$/ton$_{CO_2}$, but they foresee a significant cost reduction in next years thanks to economies of scale and learning-by-doing mechanisms (down to 50 \$/ton). Other materials are being tested to capture CO_2 from the atmosphere, especially solid sorbents, but they have not been deployed at demonstration scale, so that their real potential is still uncertain and not supported by detailed techno-economic assessment: this is the case for artificial trees developed by Lackner. For this reason, they have not been included in the modeling exercise of this research. It is likely that new designs will be developed in the future, with further cost reduction potential, as this research field is gaining more and more attention.

Results from the energy system model TIAM show that Negative Emission Technologies appear essential to reach stringent mitigation targets reducing the burden of deep decarbonization for the whole society: indeed, trying to keep the temperature increase below 2°C and 1.5°C not relying on any CDR option results in extremely high levels of carbon price (i.e. marginal abatement cost), reflecting the infeasibility of these mitigation pathways. Results suggest that an integrated portfolio including Direct Air Capture allows to reduce the mitigation effort in some sectors which are difficult to be decarbonized or with high energy intensity, such as transportation and industrial one, with less drastic decarbonization to be realized in next decades (see Figure 1a). In particular it is interesting to underline how a technology which is likely to be deployed only later in the century can have an impact in the short term, mainly looking at the energy mix, thus influencing future investment decision.

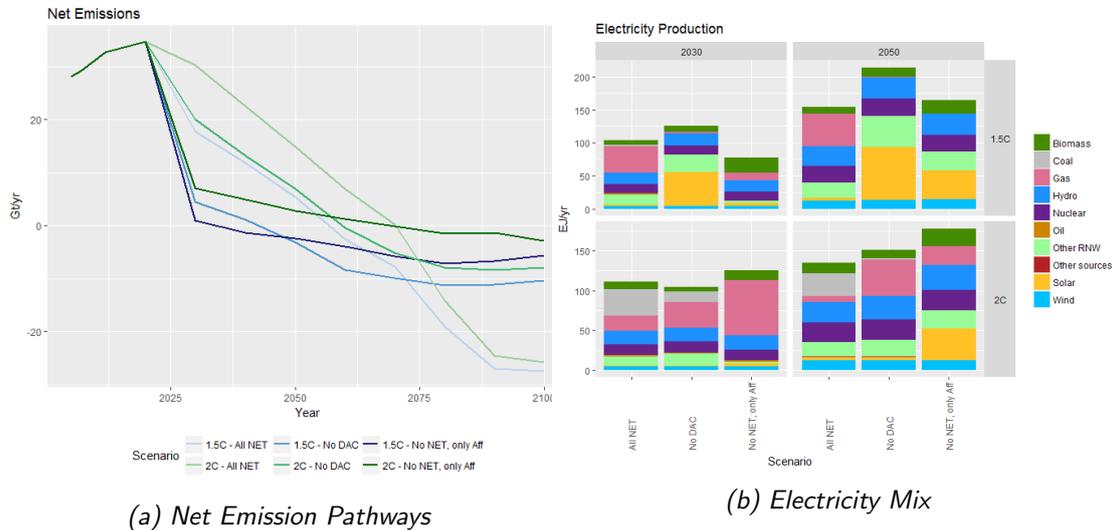(a) Net Emission Pathways

(b) Electricity Mix

Figure 1: Impact of DAC deployment in 1.5 and 2°C scenarios: net emissions and energy system.

According to model results, DAC will become a competitive mitigation option only in the second half of the century, when its capture capacity will reach a Gton scale. Nevertheless, DAC future deployment affects the electricity mix already in the mid-term (see Figure 1b), with fossil fuel still playing a role in electricity generation up to 2050 and a lower amount of intermittent renewable generation being required than if DAC is not available. In particular, considering a 2°C target, the phase-out from coal-based power plants is delayed after 2050, while the impact of DAC technologies become even more important with a stringent mitigation target. Comparing 1.5°C scenarios, it can be seen that not only the overall electricity demand is decreased of more than 25% due to a lower need for electrification in transport and industrial sector, but also the energy mix is much different when a full integrated portfolio of NET option is deployed, with natural gas power plants still playing a role up to 2050 and a reduced share of solar generation: indeed, when DAC option is not available, solar represents 40% of total generation already in 2030, with renewable sources accounting for more than 75% of the total.

The level of DAC deployed depends mainly by the overall cap applied to CDR options: while BECCS is limited to a capture rate around 10 Gt/yr by bioenergy availability, DAC takes the remaining potential available, as no other external limiting factor are applied. Considering also the results from previous modeling exercises and expert elicitation, this is likely to be between 20 and 35 Gt/yr. Among the different DAC options considered in the modeling exercise, the one based on solid amine sorbents (DAC2) is generally deployed earlier in time, while plants employing hydroxide solutions (DAC1) becomes competitive later in the century, when larger amount of natural gas are available at cheap prices to fuel them, as the role of fossil-fuel based electricity generation is strongly reduced. Moreover, DAC1 is the technology mostly affected by the presence of other competitive NET options, due to the higher energy requirements. It is interesting to notice the regional characterization of different DAC technologies, related to the different cost of commodities, with DAC1 generally installed usually in Western Europe, China and Russian area.
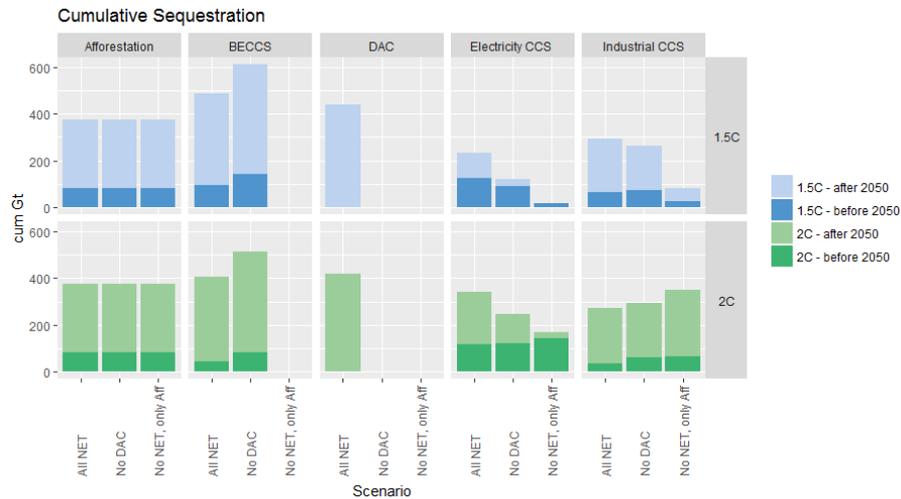

Figure 2: Cumulative sequestration of different mitigation options, in 1.5 and 2° C scenarios.

Considering the main alternative represented by BECCS plants, it can be concluded that there is not a real competition between these two options, but they needs to be developed in conjunction. On the one hand, DAC technologies are energy-intensive, requiring up to 45 EJ/yr of electricity (around 10% of global production) and 180 EJ/yr of heat to be operated, both in form of natural gas (for DAC1) and waste heat (for DAC2). On the other one, BECCS is limited to capturing around 10 Gt/yr by the amount of bioenergy that can be supplied and its sustainability: indeed, deploying DAC together with other CDR options could potentially reduce both land and water needs of about the 8 and 5% respectively. Therefore, both approaches need to be combined when ambitious climate target are set, reducing the risk and the economic impacts of relying on one single mitigation strategy.

When investigating the different sequestration options available and their timing, it can be seen from Figure 2 that the availability of DAC increases the role of CCS in the electricity sector in the second half of the century, in order to provide the required energy to operate these plants with carbon-free sources. At the same time, carbon capture in the electricity sector appears to be reduced in the short term, as DAC allow to offset emissions from industry and transport sectors, requiring less drastic decarbonization in next decades. A huge impact can be seen also in the cumulative sequestration achieved by BECCS plants, both in the short and in the long term.

Parameter Uncertainty and Sensitivity Analysis
Consider the uncertainty related to *energy and cost parameters* in the literature, it can be generally said that their influence in determining the overall capture rate of DAC plants is quite limited as the model tends to install this technology as a backstop solution to meet the climate target imposed. Differently, the most binding constraints are the one regarding growth rate and diffusion extents, as already highlighted by previous modeling exercises [7, 8]. The main obstacle for the diffusion of DAC1 are the energy requirements, while for DAC2 it is related to cost estimates. Indeed, there is higher potential for cost reduction related to amine-based technology as the design of plants can be developed and optimized for this application. Differently, for DAC1 the majority of equipment needed comes from well-known processes, as the pulp and paper industry, so that it may decrease cost more rapidly but with a limited reduction in time, that means a higher floor cost eventually reached.

The annual *growth constraint* does not affect much the extent to which DAC is deployed in 2100, being always around 20 Gt/yr out of 35Gt/yr of CDR, but the impact is more evident on the deployment during previous decades, especially around 2070-2080. This appears clearly looking at 1.5°C results, while with a less stringent target cost parameters are more relevant.
Changing the *maximum capacity* allowed for NET options, moving from 35 to 50 Gt/yr BECCS does not result much influenced, as it is limited by the availability of bioenergy, while DAC is able to capture around 12 Gt/yr more.

The model *time discount rate* determines inter-generational preferences and how mitigation effort is spread across the century. In particular, with a 2°C target and a full NET portfolio, a smaller discounting leads to a drastic decarbonization of the entire system earlier in the century, while limiting the deployment of DAC to 8 Gt/yr. Differently, with only DAC as a NET, the model still relies on its massive deployment to meet the imposed carbon budget. When moving to a stringent mitigation target (1.5°C), the role of DAC is even more essential to reduce the costs associated: a lower discount rate is not able to reduce the amount of DAC needed to meet the budget. Also BECCS deployment appears to be much less influenced by different discount rate than in 2°C scenarios, arguing that with a more stringent target the alternatives to achieve a net removal of carbon dioxide from the atmosphere needs to be deployed at their maximum potential. This suggests

that DAC technologies will be needed more if mitigation is being delayed in time, while more drastic short-term efforts could reduce the need for massive deployment of this technology and corresponding higher mitigation cost. Nevertheless, even when drastic emission reduction is applied in the first decades, DAC will still play a role in the second half of the century to reduce the impact of mitigation on the energy system.

With a reduced *CO₂ storage availability*, priority is given to DAC plants with respect to other sequestration options, as its role cannot be substituted by other technologies to tackle decentralized emissions, resulting in less BECCS and CCS in both electricity and industrial sector. Similar trends can be found with both mitigation targets. Moreover, while in some countries, moving from high to median storage potential estimate, BECCS captured capacity is being replaced by DAC plants (this is the case in China and Russian region), in most of the regions the amount of DAC is reduced as it is concentrated in the few countries that hold consistent storage resources and are characterized by lower commodity price. This means that in the base scenario the regional cost of energy commodities (heat, natural gas and electricity) is not the main factor influencing DAC deployment, while it becomes relevant when storage is a limiting factor, together with the availability of storage itself. Indeed, it should be noted that the amount of CO_2 sequestered by CDR in China does not increase much in low storage scenarios, even if it is one of the region that holds a significant fraction of the global potential, as it does not result economically convenient due to the cost of energy in these countries (note that in China mainly DAC2 is installed). Considering a limited biomass potential due to competition with land and other sustainability goals, it can be seen that the lower the deployment of BECCS as a carbon removal option, the more important the role assigned to DAC plants to achieve the imposed target, increasing its capacity of about 5 Gt/yr.

Future Work

All the results discussed so far confirm the urgency of further research on Direct Air Capture technologies, both from a technical perspective, to find the most suitable sorbent materials and optimize plant designs, and from a modeling perspective, to assess its role in future mitigation pathways. Considering this second aspect, results coming from different IAMs should be compared to check the robustness of future scenarios for DAC deployment across a wide range of model structure and functional form, given the influence that expansion constraints have on the capture rate from DAC plants. The modeling assumptions developed for this research work will be implemented in next months in other Integrated Assessment Models, namely WITCH and IMAGE, before submitting these results in a high-impact journal paper, to inform policy makers based on a solid inter-model comparison exercise.
Moreover, these research outcomes will be presented in November at the Integrated Assessment Modeling Consortium (IAMC) Meeting, as a contribution to the discussion on "Deep Mitigation Pathways".

Contents

Acronyms

APS American Physical Society

ASU Air Separation Unit

BECCS Bio Energy with Capture and Storage

CCS Carbon Capture and Sequestration

CHP Combined Heat and Power

CDR Carbon Dioxide Removal

CE Carbon Engineering (company)

CHP Combined Heat and Power

COP Conference of the Parties

CSP Concentrated Solar Power

DAC Direct Air Capture

ECBM Enhanced Coal Bed Methane recovery

EE Expert Elicitation

EOR Enhanced Oil Recovery

ETL Endogenous Technical Learning

ETSAP Energy Technology Systems Analysis Program

GHGs GreenHouse Gases

GT Global Thermostat (company)

GWP Global Warming Potential

IAMs Integrated Assessment Models

IEA International Energy Agency

INDCs Intended Nationally Determined Contributions

IPCC Intergovernmental Panel on Climate Change

LBD Learning-by-Doing

MARKAL MARKet ALLocatio

MOFs Metal Organic Frameworks

NETs Negative Emission Technologies

PEI Poly-Ethylenimine

TIAM TIMES Integrated Assessment Model

TSA Temperature Swing Adsorption

TVS Temperature-Vacuum Swing

TPES Total Primary Energy Supply

Chapter 1

Introduction and Motivation

1.1 Climate Change Framework

It is widely recognized that climate change is one of the main challenges of the 21st century. The correlation between increasing temperatures and Greenhouse Gas (GHG) emissions caused by human activity has been agreed on by the scientific community [2]: energy production and consumption, as well as transport, industry and agricultural sectors are the main sources for GHGs. The Intergovernmental Panel on Climate Change (IPCC) reports that an increasing temperatures may cause irreversible damages in terms of rising sea level, extreme weather events, loss of biodiversity and ocean acidification. These will affect the entire Earth ecosystem and all people around the globe to different extent, being likely to trigger migration movements and conflicts for water and land use in the future, with negative impacts on national economies.

In order to avoid the adverse impacts due to anthropogenic emissions, there are two approaches: mitigation and adaptation. The former one refers to actions that decrease the scale of climate changes, either reducing GHG emissions or enhancing the climate system's capacity to absorb such gases, while the latter one requires modifying habits to minimize impact on people and economies.

Recent estimates of current emissions are about 39 Gt CO_2/yr, out of which 36 Gt come from fossil fuel combustion and cement production and about 3 Gt from land use change. Less than half of current and historical anthropogenic CO_2 emissions remain in the atmosphere, while the remainder (18 Gt CO_2/yr) is being taken up by the ocean and the terrestrial biosphere, representing a natural carbon removal that moderates the impacts of human emissions on the global climate. Human activities and are altering the the natural carbon cycle as emission rates are higher than the uptake achieved by natural sinks, such as oceans and forests.

An additional challenge is the increasing demand for energy of the growing modern society. Efforts by industrialized countries to cut their emissions through electrification and the use of renewable energy sources have been more than offset by the growth in energy demand in developing nations, which is largely being supplied by fossil fuels.

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Even if scientific evidence is gaining wider consensus, the international political commitment to reduce carbon dioxide emissions proved to be slow and weak up to now. In the latest years, though, some steps forward have been made. The Conference of the Parties (COP21) held in Paris back in 2015 represented a milestone: for the first time, 197 countries agreed on concrete targets for emission reduction and policies to keep the temperature increase well below 2°C, enabling a transition towards a low-carbon world, through both mitigation and adaptation. These proposals were confirmed one year later in Marrakesh at the COP22 where countries ratified the Intended Nationally Determined Contributions (INDCs)[1].

Scenario analyses suggest that both 1.5°C [9, 10] and 2°C targets [11] are technically and economically feasible, but it remains uncertain whether future emissions will decline fast enough to meet the requirements of low temperature targets, while trying to achieve other sustainability targets (e.g. biodiversity conservation) and development goals (e.g.water and food security). Conventional mitigation, that is a reduction in CO_2 emissions, may prove not to be enough and it would be probably needed to go further, achieving negative emissions. Indeed, Negative Emission Technologies (NETs) may play a crucial role as they allow to offset emissions coming from different regions and different time periods, potentially reducing the burden of mitigation.

1.1.1 The Need for Negative Emission Technologies

Small progress have been made in global GHG mitigation over the last 20 years, adding pressure to the urgency to meet stringent climate mitigation targets.

As the temperature increase can be considered in first order approximation linearly related to cumulative CO_2 emissions [1], climate change impacts can be mitigated also by minimizing historic emissions and not only adjusting future ones [2]. This has generated interest in the potential for large-scale greenhouse gas removal from the atmosphere via Negative Emissions Technologies (NETs), as a way to reduce the carbon stock inherited by previous decades. At the same time, they can offset current emissions from sources which are difficult to mitigate directly, such as the transportation sector. Without NET, a rapid and massive expansion of intermittent renewable sources is needed to meet Paris agreement target in 2050 and further keep temperature increase below 2°C by end of century, with an impact on grid security, storage and additional cost for the entire system.

Studies on Negative Emission Technologies (NETs), also referred to as Carbon Dioxide Removal (CDR), has been conducted for almost two decades, but the topic has received more attention since the IPCC's Fifth Assessment Report (AR5) was published in 2013 [2], and will gain even more relevance as research is focusing on strategies to meet stringent mitigation targets, limiting the temperature increase to 1.5°C, as it will be investigated in the new IPCC report issued next November. Within

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[1]Up to now, 179 out of 197 countries have ratified the Paris Agreement.

the IPCC AR5, most of the mitigation pathways investigated apply global Negative Emission Technologies in the second half of the century to keep global temperature increase below $2°C$[2].

Carbon Dioxide Removal (CDR) options should be viewed as part of a wider mitigation portfolio, and not as an alternative to deep decarbonization and emission reduction in the short term: still lots of uncertainties are related to NET in terms of costs, growth rate and infrastructure needed, so that relying too much on their potential would increase the risk to postpone mitigation efforts, ultimately not achieving the target. Uncertainties are related mainly to the global capture potential of emerging and future NETs, the sustainability and the cost of achieving large-scale deployment, the carbon-climate feedback of entering a carbon-negative world and the socio-institutional barriers, including governance and public acceptance of new technologies [10].
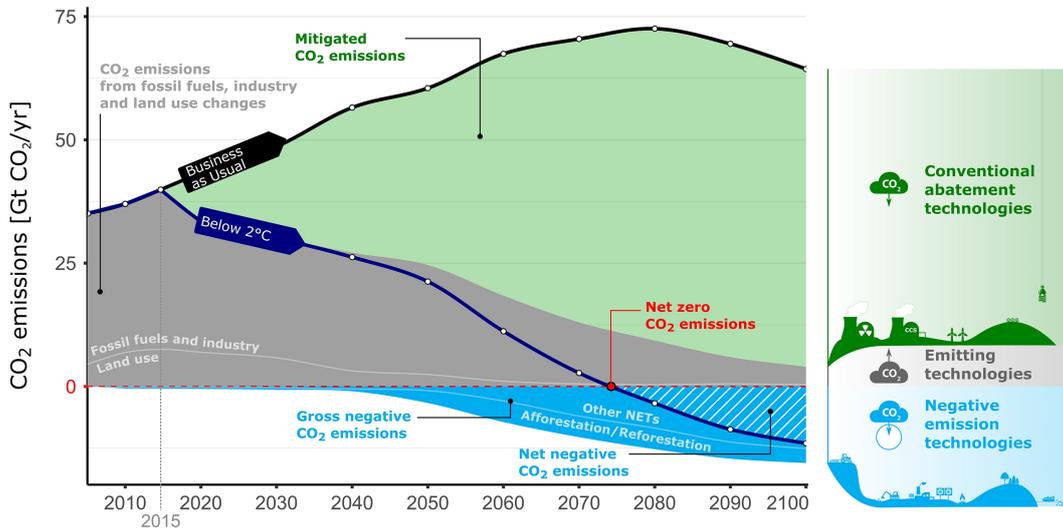
Figure 1.1: The role of negative emissions in keeping global warming below 2° C. Cumulative gross negative emissions are represented by the blue area [12].

Indicative of the current lack of commitment to NETs is their complete absence in any of the Intended National Determined Commitments (INDCs) submitted in support of the Paris Climate Agreement. At the same time, investments from the private sector are too low compared to what would be needed in the short term [13]. While renewable energy is now an attractive option, investing in CCS, NETs, or other large-scale technologies is seen as a high political and economic risk, even if the mitigation pathways to 2100 excluding NETs result to be substantially more expensive than the ones including them [11, 14]. This underlines the need to investigate further the role of Carbon Dioxide Removal technologies as part of an integrated and diversified portfolio of mitigation strategies, while informing policy-makers about the

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[2]out of 116 scenarios included in the report, 101 included NET as a strategy to achieve the target, that is the 87%

potential in the future. Moreover, negative emissions could help to offset emissions from countries that might not participate in reduction efforts or have less capacity to do so, opening new perspective on global climate management.

Direct Air Capture (DAC) is one of those technologies that allow to reach negative emissions, as it removes carbon dioxide from the atmosphere through chemical reactions. DAC would compete - or might be deployed in parallel - with two terrestrial biological CDR strategies, such as afforestation and CO_2 capture from bioenergy facilities, namely BECCS. The advantage of this engineered carbon removal is that it allows to offset emissions coming from distributed sources, such as the road transport and the aviation sector, while reducing the amount of land and water needed to achieve large-scale negative emission compared to the biological options.
Carbon removal have been already assessed and implemented by many Integrated Assessment Models (IAMs) studies, focusing on biological technologies as the most-favorable to be deployed in short term, but there is lot of discussion and concerns about their land footprint and possible competition with food and water supply, conflicting with other sustainability goals [15].

1.2   Research Questions and Methodology

Many studies that have evaluated the amount of negative emissions achievable through the deployment of BECCS or DAC, considered them in individually [4, 5, 6], not focusing on their mutual interaction and the role of these technologies within the broader energy system. Carbon capture, conversion, storage and/or utilization add complexity to the mitigation framework, and a number of questions about rates, locations, amounts, costs, infrastructure, still needs to be addressed with a systemic approach, with priority attention to be dedicated on their sustainable and available potential, both in term of technology development and market deployment [16].

Starting from a previous thesis work [8] about a multi-model assessment on the role of Direct Air Capture and considering the ongoing scientific debate on the potential of these technologies, this research work aims to couple a techno-economic analysis of the capture process itself, with an evaluation of its role in future mitigation scenarios through the use of Integrated Assessment Models (IAMs). From previous IAM-based works [4, 8], DAC deployment results very sensitive to the feasibility constraint applied in term of expansion rates, as it represents a backstop technology, that caps the marginal cost of abating GHG emissions and act as an anchor for climate policy. Therefore, boundary values and ranges of costs for DAC need to be chosen carefully, as any model will go straight to this kind of technology as soon as it becomes competitive, leading to unrealistic overshooting and exaggerated penetration rates.

As this is the final work at the end of five years of studying about the energy sector, I tried to combine a technical perspective on the design of DAC plants, considering the impact of different technical aspects, energy consumption, and costs of individual components, with a policy-oriented one, assessing the feasibility of future deployment scenarios and the impacts on the broad energy system, while conducting extensive sensitivity analyses with respect to a number of the key parameters in order to ensure the robustness of our results. The goal of the analysis, therefore, is not to provide specific levels of the policy variables, but rather to assess the optimal portfolios and identify the crucial parameters affecting it.

By understanding the effect of different policies on the deployment of DAC technologies, the final aim is to help governments and firms decide among different investment strategies for capture technologies.

The research questions I tried to address with my work are the following:

1. Which are the technology options available in order to capture CO_2 directly from the air? How can they be characterized in terms of costs and energy requirements?

2. Which could be the physical scale of DAC deployment? Which could be its impact on the energy system and in term of land/water/material use compared to other NET options?

3. Which is the role of DAC as part of the Negative Emission Technology (NET) portfolio on mitigation pathways with stringent climate targets, consistent with 2°C/1.5°C increase in temperature? In particular, which is the relative role of DAC and Bio Energy with Capture and Storage (BECCS), and how the deployment of the former one can affect the role played by bio-energy and other sequestration options?

4. To which extent uncertainties on cost and energy estimates, deployment constraints, discount rate or storage potential can affect the role of DAC in future mitigation scenarios?

1.2.1 Research Methodology

Considerations about the future role for DAC and its future cost are constrained by the scarcity of experimental results and detailed engineering assessments of these systems, as few pilot-scale DAC plants have been deployed. Nonetheless, DAC has entered policy discussions and it is likely to gain increasing attention in next years.

The main tool used in my research to assess the role of DAC are Integrated Assessment Models (IAMs), which are designed to investigate possible long term energy futures, based on the interaction of different dimensions. Indeed, they couple a representation of the economic and energy systems with a climate model, describing GHG emissions and their impact on temperature, so to understand the basic mechanism involved. In particular, I have been working with TIMES Integrated Assessment

Model (TIAM), maintained by the Grantham Institute at Imperial College London.

As an accurate modeling with a strong engineering basis is needed in the climate field to understand the impact of new technology options in a century-long perspective, my thesis work tries to couple a techno-economic assessment of different design proposed in the literature for capturing CO_2 from the air, with expert elicitation and results from previous modeling exercises to understand possible deployment pathways. In order to deal with the uncertainties related to this technology, modeling results are considered under a broad range of sensitivity analysis, regarding energy and cost assumptions, as well as growth constraint, discount rate applied and storage availability. In particular, my research work was structured according to the following steps:

1. *Literature Review*: collecting information on both technical and economic specifications for DAC systems, so to provide the background for an accurate modeling. Moreover, historical deployment rates and limiting factors for the diffusionn of 'competing' NETs, mainly BECCS, have been investigated.

2. *Expert Elicitation*: in order to overcome the scarcity of clear technical data for DAC plants, an expert elicitation was designed to understand possible future development for these technologies based on the perspective of people actively researching in this field

3. *Implementation* of DAC technology options within the TIAM framework, including them in the wider Reference Energy System. Given the technical granularity of this model, this step results key to assess the impact of DAC on the overall energy system.

4. *Scenario Design and Robustness Analysis*: once DAC plants have been integrated in TIAM, at first diagnostic runs have been carried on, before moving to scenarios with different mitigation targets (i.e. carbon budget compatible with 2/1.5°C increase in temperature). Sensitivity analysis was then designed to assess the impact of different parameter uncertainties and modeler's choices on the overall deployment of the examined technologies.

5. *Analysis and Discussion of Results*, coming both from the main runs as well as from the robustness analysis. Key findings are then presented with a policy-making perspective.

1.2.2 Thesis Structure

The structure of the thesis report is the following:

- In Chapter 2 the current state of the art for DAC options is discussed, combining technical designs found in the literature with first pilot plants and the perspective of companies behind them. Both the energy requirements and the cost estimates are presented.

- In Chapter 3 a general description of IAMs is provided, with a specific focus on the model TIAM used in this work. Moreover, it is discussed how experts' opinions have been included in this research work through the development of an Expert Elicitation questionnaire.

- In Chapter 4 the implementation of DAC technologies within the TIAM modeling framework is discussed. Both techno-economic parameters and growth constraints will be considered, as well as the interaction of DAC with the overall energy system, given the high level of details available in technology-rich models as TIAM.

- In Chapter 5 the main results on the possible role of DAC as part of a NET portfolio are presented, considering mitigation pathways consistent with both 2°C and 1.5°C increase in temperature by the end of the century.

- In Chapter 6 the extensive sensitivity analysis is presented, including results and discussion of the impact of different parameters on the deployment of DAC and other mitigation strategies throughout the century.

- In Chapter 7 the key findings are discussed with a policy-making perspective, and some suggestions are drawn for future research works needed to assess the impact that these technologies may have in the future.

- In the Appendix further information about TIAM model and DAC implementation within it are presented, together with the full questionnaire designed for the expert elicitation.

Chapter 2

Direct Air Capture Technologies

DAC is a rapidly growing environmental technology and it is gaining more and more attention in the academic field, with an increasing number of research focusing on developing materials and processes for this applications, as well as few start-up companies pushing this technology from lab to demonstration and pilot scale.

With demonstration plants being installed, both in Canada by Carbon Engineering back in 2015 and in Switzerland by Climeworks just in 2017, Direct Air Capture is getting closer to the market deployment but there is not yet a convergence on the technical design. Nevertheless, first commercial plants represent a very important step, proving to the world outside of the laboratory that DAC can actually work.
In next Sections 2.3 to 2.5, the main technology options that can be found in the literature will be discussed, providing a description of the proposed plant designs and presenting private manufacturers and start-ups which are implementing some of them at a demonstration level[1]. At the end, energy requirements and cost estimates are presented, in Sections 2.7 and 2.8.
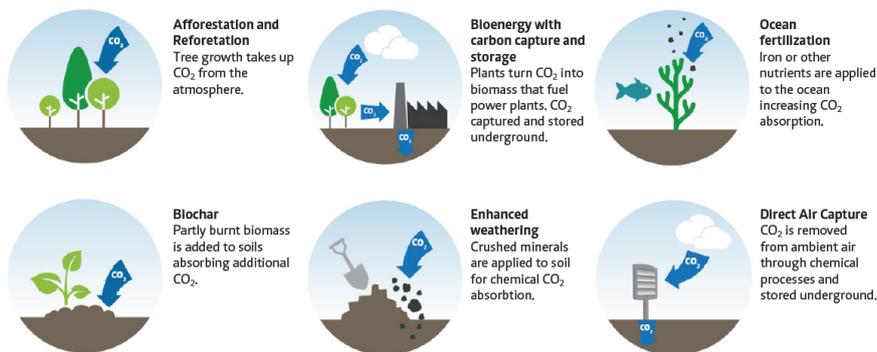

Figure 2.1: Different CDR options [18]

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[1]Note that all the companies analyzed were named as finalists in the Virgin Earth Challenge launched in 2007 [17]

2.1 General Framework: NET Options

Some CDR approaches are meant to amplify the rates of processes that are already occurring as part of the natural carbon cycle (i.e. land management, ocean iron fertilization, accelerated weathering), while others involve capturing CO_2 from the atmosphere, concentrating and storing it (i.e. BECCS and DAC).

According to the classification by the Royal Society [19], there are two *biological terrestrial CDR strategies*, which build on the natural cycle of plants taking CO_2 from atmosphere to be used during photosynthesis processes.

1. *Afforestation and Land Use*: as growing trees bind emissions, for each hectare of reforested land about 500 ton CO_2 can be removed from the atmosphere. In addition to it, it mat bring other environmental benefits, such as water management and purification, and protection of biodiversity. This strategy come at relatively low costs, however the main drawback is the land-intensity [20]. Another option could be reducing the rate of deforestation.

2. *Biological Energy with Carbon Storage (BECCS)*: a power plant based on biomass is coupled with a capture unit for the CO_2 released during combustion. Considering the entire life-cycle of the fuel, there are two capture steps, firstly during the growth of biomass due to photosynthesis, and secondly at the power plant after combustion, with an overall net negative balance. At the same time useful heat, power, fuels, and synthetic gas for chemicals and fertilizer can be produced without fossil fuels. Due to the large consumption of land, BECCS could interfere with other goals, such as food security, therefore social acceptance results quite low.

In addition, other *terrestrial chemical CDR strategies* are:

1. *Direct Air Capture (DAC)*, using chemicals such as amine or strong base (e.g.sodium hydroxide) solutions to absorb the CO_2 from the atmosphere, then sending it to storage sites. This option requires little land, but the energy needed would count to roughly a third of the world's energy demand [18].

2. *Acceleration of weathering*: weathering of minerals is a natural chemical process removing CO_2 from the atmosphere on a multi-thousand-year time scale. Accelerated weathering strategy is based on grinding and spreading rocks, both on land or the ocean, that naturally absorb carbon dioxide, thus increasing their surface area and the rate of the reaction

3. *Biochar*, a form of charcoal, made from biomass through pyrolisis, thus sequestering carbon dioxide while increasing soil fertility and agricultural productivity. Its potential is quite limited and still under investigation.

Additional removal option could be *Ocean-based CDR* (ocean biological fertilization, or chemical alkalination) and *Solar Radiation Management*, but the uncertainties about real effectiveness and risks involved are still pretty high. Generally, CDR

technologies are affected by uncertainties, about their technical potential, economic costs, adverse side effects and sustainability implications, and political feasibility, related to policy-making and governmental support to achieve a net carbon dioxide removal [6]. On the one hand, R&D investments can improve processes for carbon dioxide removal and sequestration, in particular to minimize energy and materials consumption, lowering the related costs. On the other hand, widespread CDR deployment would likely occur in a policy environment in which there are limits on CO_2 emissions or a price is imposed on them, as CDR will compete directly with other mitigation strategies on a cost basis.

Carbon Capture and Sequestration (CCS) from power plants prevents CO_2 emissions but does not remove carbon dioxide from the atmosphere, therefore it can not be considered a CDR option, even if it holds some similarities with air capture processes. The biggest difference between DAC and BECCS as CDR option is that the former requires an energy input, while the latter one can produce electricity while sequestering.
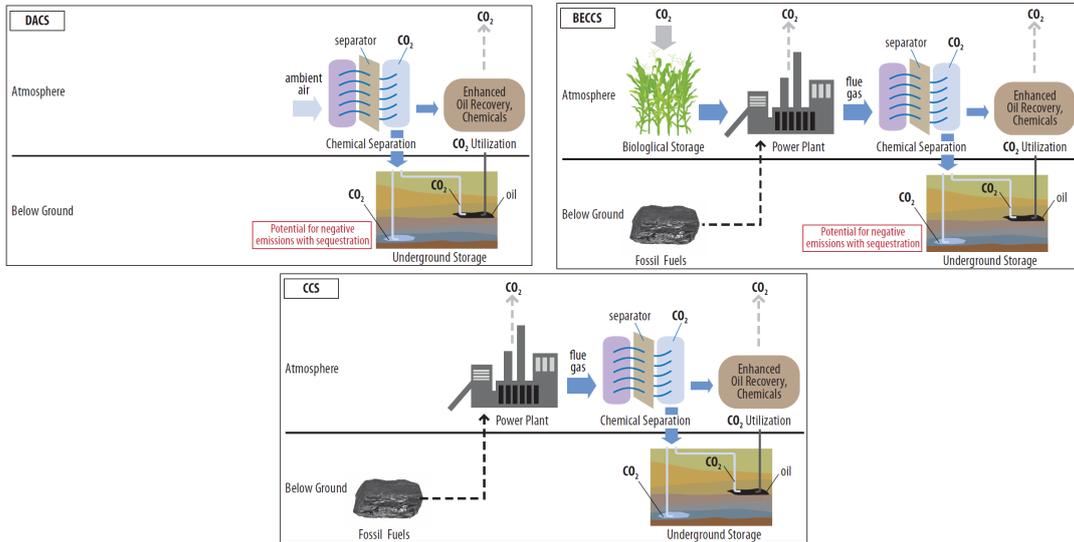
Figure 2.2: Different sequestration strategies: DAC, BECCS and CCS [21].

2.2 Direct Air Capture

The capture of CO_2 from ambient air was commercialized in the 1950s as a pre-treatment for cryogenic air separation, while in the 1960s it was proposed as a feedstock for the production of hydrocarbon fuels using mobile nuclear power plants [22]. In the 1990s, Klaus Lackner [23] was the first to explore the application of large-scale carbon capture as a mitigation tool to reduce the climate impact of anthropogenic emissions, and this is now commonly referred to as Direct Air Capture. Later in 2011, the report by the American Physical Society [20] represented the first detailed technical and economic assessment of a Direct Air Capture plant based on hydroxide solutions, being still one of the main reference in this research field.

The literature is still extremely fragmented on the technology options available to realize such process: only in the last years it has been started to refer to this category of technology as DAC in a consistent way. This lack of identity do have an impact also in the academic, political and public sphere, bringing not sufficient attention to this topic and to its role in reaching ambitious climate goals. This is translated in a lack of integrated research programs to support its development.

2.2.1 General Characteristics

DAC may play a role in reducing decentralized CO_2 emissions that prove expensive to be reduced in other ways, such as the ones from buildings and vehicles (ships, planes and the transport sector in general), or to offset emissions from energy-intensive sectors, difficult to be decarbonized, such as steel and cement manufacture.

Being the concentration of CO_2 in the air so dilute and the partial pressure so low, the only practical option is to use chemical sorbents, while for flue-gas capture both physi-sorbent and chemi-sorbent materials are applied.
Generally, the process of CO_2 capture from air holds some similarities with post-combustion scrubbing technologies from power plants (CCS), both involving an absorbent and consisting of three main steps [24]:

- *Contacting Ambient Air*: it requires a physical structure channeling the air to the sorbent surfaces. Air can be driven by a machinery (e.g. fans) or by a natural flow due to ambient conditions (e.g. natural wind, thermal convection, or wind-driven pressure gradients). The use of fans is limited to low velocities to minimize pressure drops and energy consumption.

- *Absorption or Adsorption* with a liquid or solid sorbent. It is an exothermic reaction, thus releasing a certain amount of energy in form of heat

- *Sorbent Regeneration* to release the carbon captured during the previous step. It is a very energy-intensive step (i.e. endothermic reaction), with the energy consumption being proportional to the mass of the captured carbon dioxide.

Capturing CO_2 directly from air involves important challenges with respect to other Negative Emission Technologies (NETs) or to traditional CCS, mainly related to the low concentrations of carbon dioxide at atmospheric pressure, the high energy requirements, the limited range of operating temperature and the presence of moisture that may affect DAC plants performances [25]. Therefore, the key aspects to be addressed are choice of an appropriate absorbing material, as its properties determine the entire energy balance and the regeneration system, the regeneration system and the contactor design, that should put in contact a wide area of absorbing material with the flow of air from the atmosphere, balancing the amount of sorbent and the energy penalties due to pressure drops [24].

Most of the technologies proposed for DAC rely on batch processes, that separate collection and regeneration in different steps, therefore distinct units of operations.

Compared to other sequestration options, capturing CO_2 directly from the air allows a number of advantages, that would be key in determining its large-scale deployment in the future:

- It allows to address *distributed emissions* coming from home/office heating and cooling as well as the transportation and aviation sector: these account for almost the 50% of the total. Indeed, collecting carbon dioxide from small burning units at the source often results difficult and not economical with large CCS facilities. Note that DAC is not the only way to abate these emission sources, but the cost of the alternatives, such as electrification of transport and building sectors or an increased the use of biomass, are expected to be considerably higher than the current estimates for DAC [4].

- Being independent from the CO_2 point source means that the capture unit could be placed anywhere, offering a significant *location flexibility*. As a consequence, transportation costs and risks associated with moving the captured CO_2 to the sequestration sites can be reduced. This would also increase the social acceptability for this type of intervention, as long pipelines can be avoided.

- DAC plants can scale rapidly through *modularity*, as they can be optimally sized to suit geological sequestration sites and the available technology, with no need to be coupled with (and adapted to) existing systems and plants.

- Ambient air generally has a much *lower concentration of contaminants* such as NOx, SOx and particulates with respect to flue gases: these normally represent a major cause of degradation and reduced performance in flue gas capture processes.

Differences with Traditional CCS

Even if the ultimate effect is to separate CO_2 from a gas stream, Direct Air Capture (DAC) and flue gas capture (i.e. traditional Carbon Capture and Storage - CCS) are distinct technologies, so that they are not considered them as exclusive options, but a parallel deployment can be conceived. The main difference is that DAC could be used not only to prevent CO_2 concentration from increasing but even to lower it, offsetting previous emissions and achieving a net negative carbon balance. Negative emissions can be achieved also when traditional CCS is coupled with biomass utilization as explained before, but the main limitation will be related to land availability and its competition with food and energy security [26].

Other distinctions among these two options derive from the different concentration in the stream to be treated [21]:

- As CO_2 in the air is 100 to 300 times more dilute than in combustion gases[2], the capture process is more energy-intensive: the minimum amount of energy required is about 3 times the one to capture CO_2 from flue gases.

- Due to the different dilution, absorption unit for DAC is likely to be large in terms of cross-sectional area, but very shallow to limit pressure drops, while CCS unit is tall and potentially thin. As a consequence, the footprint in term of land use is much different, with CCS requiring more land to capture the same amount. These have an impact on capital costs, as they generally scale with land area.

- Concentrated emissions from power plants are easier to target due to their higher concentrations of carbon dioxide, the known and controlled exhaust qualities and flow, and the presence of a responsible party to quantify emissions [27]

2.2.2 Theoretical Energy Requirements

From a theoretical perspective, the free energy required to separate 1 mole of CO_2 from a gas mixture is given by the following equation:

$$\Delta G = RTln\left(\frac{P}{P_0}\right),$$

where, R is the universal gas constant, T is the temperature of the gas, P is the partial pressure of the CO_2 at the exit of the scrubber, and P_0 is the pressure of the gas to be separated, that is the pressure of ambient air. Considering ambient conditions($T = 300$ K, $P_0 = 10^5$ Pa), the free energy of absorption from air turns to be at least $\Delta G = 20$ kJ/mol (0.45 GJ/ton CO_2). In practice, sorbent binding energies tend to be several times larger than the free energy change, typically well above 50 kJ/mol. A second law efficiency for different DAC processes proposed can be then computed comparing the actual energy required with the theoretical amount: generally, the realistic range for this efficiency is between 10 and 15%.

It should be noted that the binding energy scales with the concentration at the exit (i.e. the partial pressure) only in a logarithmic way, so that the effort in separating the CO_2 from the sorbent during regeneration will be very similar for both air capture and for flue gas scrubbing. This energy demand alone results much higher than the cost of contacting the air stream.

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[2]Ambient air contains around 400 ppm by volume, while the exhaust gas of natural gas and coal power plants are around 30 000 to 150 000 ppm by volume respectively.

2.2.3 Technology Classifications

Many processes have been proposed for Direct Air Capture, but only few have been tested in demonstration plants, due to the low level of technology development and the lack of consensus regarding the research and the results claimed by some manufacturers [27]. As there is still no technological convergence, estimations about the actual energy demand and economics of a large-scale plants are highly uncertain.

According to the literature, the main options are:

1. *Aqueous Solutions of Strong Bases* (Section 2.3): water solutions containing hydroxide sorbents with a strong CO_2 affinity, such as NaOH, KOH and $Ca(OH)_2$, as the ones proposed at first by the APS in 2011 [20, 28, 29, 30].
   The main company developing this approach is *Carbon Engineering* (CAN)

2. *Organic/Inorganic Solid Adsorbents* (Section 2.4): based on different solid-supported amine materials, mainly bonded to a porous support [25]
   Global Thermostat (US) demonstration plant is based on a solid tertiary amine, while *Climeworks* (CH) is developing an amine-functionalized cellulose.

3. *Other Solid Adsorbents* (Section 2.5): this category encompasses a range of different sorbents, still in the research stage with no existing demonstration plants. An ion exchange membrane, known also as "artificial tree", is being developed by Lackner [31] and the Center for Negative Emissions in Arizona, while new materials, such as Zeolites, Metal Organic Frameworks (MOFs) [32] or Alumina [33] and solid oxide [34] are being investigated.

Solid sorbents have lower energy input and operating costs, but degradation issues may arise, impacting on costs and economic life of the plant. On the other hand, while batch processes for solid sorbents require temperature, pressure or humidity to be cycled during the regeneration step, liquid sorbents offer the advantage that the contactor can operate continuously, in parallel with the regeneration step. As a consequence, the contactor can be built using cheap cooling-tower hardware, with a longer lifetimes, and a central regeneration facility can be adopted leveraging economies of scale. The main disadvantages are the cost and complexity of the regeneration system, including the high thermal needs and water losses occurring in dry environments [35].
Generally, liquid sorbents are applied in large scale plants, while solid ones allow a higher modularity, with small capture facilities.

2.3 Aqueous Solutions of Strong Base

Employing strong base solutions is the most developed and technically feasible DAC option, and extensive literature studies can be found on it, starting from the milestone review carried out by the American Physical Society (APS) back in 2011 [20].
The strong bases proposed are sodium hydroxide (NaOH) or potassium hydroxide (KOH), coupled with calcium hydroxide $Ca(OH)_2$ to facilitate the regeneration of the capture solution.

The use of calcium hydroxide $Ca(OH)_2$ was at first proposed by Lackner in 1999 for DAC application [23]: once the basic solution absorbs CO_2, it creates solid calcium carbonate $CaCO_3$ as a precipitate, which can be easily removed, dried, and regenerated in a kiln where CO_2 is released. This *calcination* recovery process is already used widely in the industry, in particular in the pulp and paper sector.
The main drawbacks of this approach are the large amount of water that can be lost through evaporation, the buildup of solid material on the equipment and the very high temperature required during the regeneration step [25]. In the last decade, researchers tried to overcome these problems by combining $Ca(OH)_2$ with other hydroxide solutions, in a double loop process.

Baciocchi [28] was the first one to propose the use of sodium hydroxide (NaOH), similarly to the Kraft process, widely used in paper industry to extract cellulose from wood. Indeed, sodium hydroxide has a sufficiently strong binding of CO_2, with the additional benefit that the carbonate formed is highly soluble in water. Even if most existing processes employ NaOH as a sorbent, potassium hydroxide (KOH) can be a viable alternative, still more expensive than NaOH. The only demonstration plant capturing CO_2 from the air with basic solutions, run by Carbon Engineering, is actually based on KOH: in the future, it will be probably easier to produce this sorbent material rather than NaOH, that requires large energy input during the synthesis process (see following discussion in Section 4.5).

Both of these methods, using calcium hydroxide alone or coupled with sodium/potassium hydroxide, involves regeneration in a kiln with a temperature around 700-900°C. To supply this high-temperature heat a fuel needs to be burnt, requiring an additional system for capturing the released CO_2 afterwards. To facilitate this second capture step, the kiln could be fed with pure oxygen, but the separation of O_2 from the atmosphere in an Air Separation Unit (ASU) involves additional electrical energy consumption, thus increasing fuel costs [25].

2.3.1 Technical Design

As said before, one of the key references in the field of DAC is the report developed by the APS [20], where a detailed energetic and economic analysis is reported. The reference plant is based on the scheme proposed by Baciocchi [28], consisting of a two-loop hydroxide-carbonate system: the first one is a NaOH - Na_2CO_3 cycle (sodium hydroxide and sodium carbonate), combined with a $Ca(OH)_2$ - $CaCO_3$ one (involving calcium carbonate, calcium oxide and calcium hydroxide).

Some criticism were raised as this design did not refer to a demonstration plant, so that cost estimates result unfounded in industry, while assumptions did not reflect the realistic potential of this technology. Nevertheless, the APS report has been heavily referenced in DAC literature. Recently, a new study [35] has been published, using data coming from the pilot plant built in Canada by the company Carbon Engineering. The plant scheme is very similar to the one proposed in the APS report, with two connected chemical loops, but potassium hydroxide KOH is being used instead of sodium one.

In next paragraph, the main steps of APS reference design are described in details.

American Physical Society Reference Design: Process Steps

The complete absorption and regeneration process involves four reaction steps, as it can be seen in Figure 2.3.
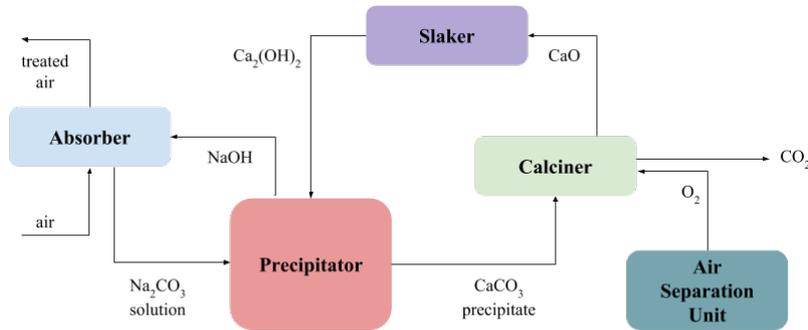
Figure 2.3: Process Steps, according to APS design [20].

Step 1: Absorber (Air Contactor)
CO_2 is captured by a solution of NaOH and converted into a solution of sodium carbonate Na_2CO_3, according to the following reaction:

$$2\ NaOH + CO_2 \rightarrow Na_2CO_3 + H_2O \quad \Delta H = -105\ kJ/mol^3 \qquad (2.1)$$

This step is exothermic, involving a reaction enthalpy ΔH equal to 105 kJ/mol: the strong binding associated with this sorbent allows a high loading of CO_2 over a wide range of operating conditions, but at the same time it requires large amount of energy for regeneration.

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[3]Note that all these values include also solvation energy, that is the energy associated to dissolving a solute in a solvent

The most common industrial method for gas-liquid absorption is to drip the solution through a tower filled with packing materials. Moving to DAC applications, one of the main challenge is the large volume of air that needs to interact with sodium hydroxide solution during this step. Different contactor designs have been proposed [36]: as it needs to have a wide cross section area due to the large amount of flow to be treated, the optimal design would be very different from conventional packed tower. Indeed, it is likely to be more similar to a trickle-bed filter used in waste-water treatments, that is a wide cylindrical basin with a rotating distributor arm, or to a taller tower with lighter packing, as power plants evaporative cooling tower or SO_2-scrubbing tower for combustion flue gas [30]. It has been highlighted that cost estimates for the contactor equipment can be largely reduced developing specific and optimized designs for DAC application, rather than applying existing industrial solutions [30, 28, 29].

Additional electric energy is required in this step to move air and NaOH solution through the contactor with fans, as the natural flow often results not sufficient.

Step 2: Precipitator (Cauticizer)

The sodium carbonate Na_2CO_3 is highly soluble, therefore a large quantity of water needs to evaporate to obtain a solid precipitate, being too energy intensive. To avoid it, Na_2CO_3 is converted to calcium carbonate $CaCO_3$ by adding calcium hydroxide $Ca(OH)_2$ slurry, according to the equation:

$$Na_2CO_3 + Ca(OH)_2 \rightarrow CaCO_3 + 2NaOH \quad \Delta H = -8 \; kJ/mol \qquad (2.2)$$

This step is slightly exothermic, but the equilibrium can be driven towards $CaCO_3$, thanks to its precipitation. With this *causticization* reaction, the NaOH solution can be regenerated and recycled back to the absorber, with a reduced energy consumption. This step is very similar to the Kraft recovery process traditionally applied in the paper industry: given the small and favorable differences of being adapted to DAC plants, a conservative estimate of the monetary and energy costs for running this component can be lifted directly from the pulp and paper sector.

Step 3: Calciner (Kiln)

The $CaCO_3$ precipitate is at first dried to remove excess water using waste heat from the kiln, then it is regenerated with a two-step process. The first one takes place in the kiln, where it is converted to CaO (quicklime), releasing CO_2 through *calcination*, according to:

$$CaCO_3 \rightarrow CaO + CO_{2 \; (gas)} \quad \Delta H = 179 \; kJ/mol \qquad (2.3)$$

This reaction is highly endothermic, requiring high-temperature heat (T > 800°C) to allow the release of CO_2 at atmospheric pressure: it is by far the most energy-intensive step due to the strong binding within $CaCO_3$ molecules. Oxygen-blown combustion can be used in a fluidized bed, so that the system will look like a hybrid of existing fluid bed calciners, currently applied in lime, cement and paper industry, and oxygen-fired coal combustion boilers with CO_2 capture. At the end of this step, CO_2 is compressed to be transported to the storage site and then sequestered.

Step 4: Slaker

The second step needed to complete calcium hydroxide regeneration takes place in the slaker, where calcium oxide CaO reacts with steam according to:

$$CaO + H_2O \rightarrow Ca(OH)_2 \quad \Delta H = -65 \ kJ/mol \tag{2.4}$$
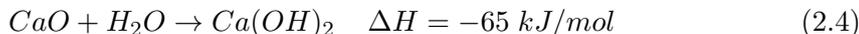
As a result, a suspension of calcium hydroxide $Ca(OH)_2$ is obtained, which is then recycled to the precipitator to close the chemical loop. The thermodynamic advantage of steam slaking over conventional water slaking used in the Kraft process is that the reaction enthalpy is released at higher temperatures.

In order to provide the required heat for the calcination step, natural gas needs to be burned, either in air or pure oxygen. The APS study is based on an oxygen-fired kiln, with a post-combustion capture system to sequester the CO_2 coming from natural gas combustion and to store it together with the one coming from air. This additional capture unit will be the same applied in oxyfuel CCS for natural-gas fired power plants, with capture efficiency usually around 95%.

The main challenges of this design are related to the fouling of the absorber with $CaCO_3$ and aerosols in the intake air, water evaporation from NaOH solution in the absorber and the loss (with consequent make-up needed) of NaOH solution entrained in the CO_2-free air leaving the absorber. Another aspect that needs to be tuned carefully is the concentration of NaOH in the solution: indeed capture is enhanced at high molarity, but it also implies an increased viscosity and a harsher environment.
To reach economies of scale for this technology option, a plant capacity of about 1 Mt_{CO_2}/yr is required, that means the system needs to process 46 000 m^3 /s of air.

2.3.2 Existing Companies and Demonstration Plants

Carbon Engineering

Carbon Engineering (CE), led by David Keith, professor of applied physics at Harvard, and funded by Bill Gates, is working on a technology combining KOH and $Ca(OH)_2$ solutions for carbon capture since 2009[4], with a double chemical loop similar to the one discussed previously (see Figure 2.4). They are running a prototype from 2011, and from October 2015 a demonstration plant is being operated in Squamish, British Columbia, with a capture capacity of 1 ton_{CO_2}/day. They claim that the first industrial-scale plant could be ready in 2020: the proposed design includes existing processes and technologies that are already widespread, as the calcium regeneration cycle similar to paper manufacturing, increasing the potential to lower the costs and to scale up quickly. The latest paper issued in June 2018 [35] provides detailed techno-

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[4]Note that the paper by Keith back in 2006 [36] proposed for the first time the process based on KOH, which is the base concept behind CE demonstration plant. Potassium hydroxide was adopted so to improve CO_2 uptake kynetics

economic estimates, based on a combination of data from vendors and from their pilot plant, along with data from the minor unit operations. These have been used as the main reference for cost and energy estimates to be implemented in TIAM model (see Section 2.8).
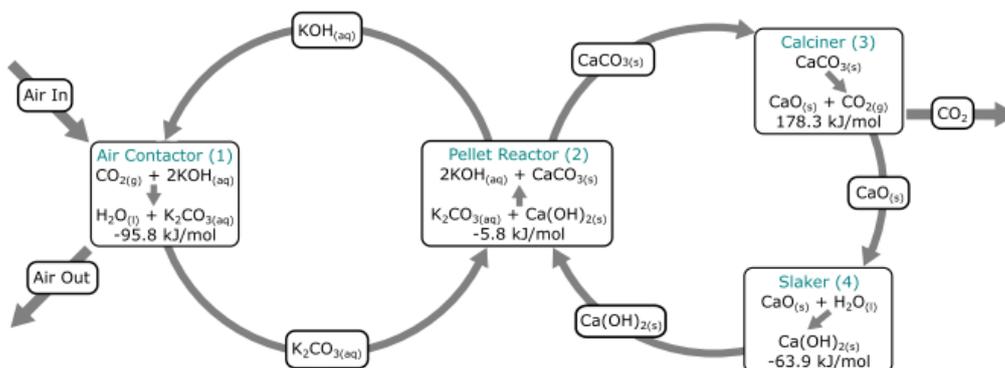

Figure 2.4: Process chemistry and thermodynamics of CE plant [35].

A key aspect that makes CE technology competitive on the market is the reduced cost for the **contactor**, resulting 4 times cheaper that APS estimate [30]. This difference is not due to costing methodology, but to a different design choice: while the APS system consists of a closed, counter-flow packed column, as it is common for a chemical scrubbing processes, CE adapt the technology used in large scale cooling towers and waste treatment, that are designed to bring very large quantities of ambient air in contact with a fluid in an efficient way. This results in an open contactor with cross-flow and a slab geometry[5], as it can be seen in Figure 2.5: the aqueous hydroxide solution flows downward through structured packing through which the air flow horizontally, with the two flows being orthogonal to each other. Structured packing is represented by PVC corrugated sheet filled with the alkali absorbing solution.
Closed systems are most commonly used to perform mass transfer on ducted, often toxic, gas streams within chemical processing facilities, while open systems are the dominant choice for ingesting large quantities of ambient air for cooling applications. Performance and development risks are associated with both designs, with open-flow systems involving higher technical risks, but also potentially reducing costs as more aligned with DAC specifications[6].

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[5]The contactor is designed to be thin along the direction of air flow compared to the overall height and length of the unit. In the demonstration plant it results 20m tall, 8m deep and 200m long, with an overall footprint of 1600 m^2 for the contactor only [35].

[6]Note that the APS report already recognized the reduced cost that may come from employing an open-flow system but discard this in the analysis due to high technical risks involved. It should be remembered that this was the first study focusing on the technical assessment of DAC plants, thus considering minimum-risk baseline technologies, while CE is building on several years of research for this specific application

The final design has benefited from a close collaboration with SPX Cooling Technologies, a leading vendor in the field of cooling towers: even if geometry and fluid chemistry are different, CE's design relies on many of the same components, including fans, structured packings, demisters, fluid distribution systems, and fiber-reinforced plastic structural components [35]. .
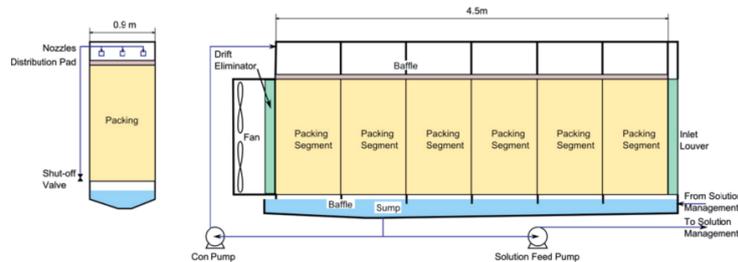

Figure 2.5: CE air contactor design, with cross flow and slab geometry [37].

Another interesting and cost-competitive aspect of CE plant is the design of a pellet reactor, instead of the traditional precipitator and the oxy-fired circulating fluidized bed calciner applied for the Kraft process in the paper industry. In the reactor, pellets of CaCO3 are suspended in a solution that flows upward, while a slurry 30% of $Ca(OH)_2$ is injected into the bottom of the reactor vessel: being able to precipitate pellets of calcium carbonate, rather than lime mud, increases performances, as pellets are washed and dried more easily, reducing energy consumption in the kiln and bringing thermal efficiencies up to 78% (compared to 39% of lime mud calciners).

One of main trade-off between capital cost (i.e. size of the contactor) and operating cost (i.e. fan power consumption) is the choice of air velocity: their optimization led to a velocity around 1.3 m/s, with an optimal capture fraction of the 75% [7].

In order to reduce loss of hydroxide solutions in liquid droplets, CE design has twofold solutions: a demister section at each face of the contactor; and operation at low-flow regime leading to smaller drop generation. a Low liquid flow rate further reduce pumping costs and equipment.
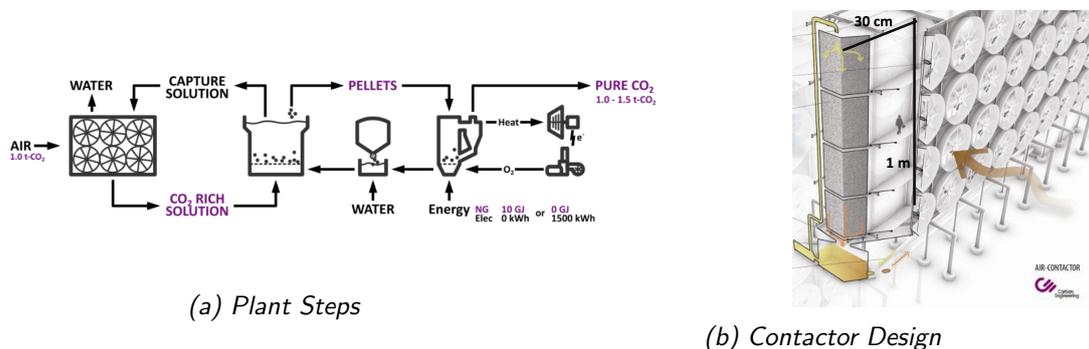

(a) Plant Steps

(b) Contactor Design

Figure 2.6: CE plant design: main process steps and air contactor.

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[7]Note that also the CO_2 capture fraction is not a specification but a design parameter resulting from an optimization trade-off on total costs

In addition to the main steps required to capture CO_2 from the air, CE demonstration plant include minor units, such as the *Power Island*, consisting of a natural gas turbine, followed by an HRSG to produce the heat and the electricity needed to drive the entire process, the *CO₂ Absorber* to capture carbon dioxide contained in gas turbine exhausts and convey it to the main air contactor, the *CO₂ Compression* setup and the cryogenic *Air Separation Unit* needed to feed pure oxygen to the calciner.

In order to launch their technology on a commercial scale, CE developed the business idea of Air-to-fuels, producing synthetic fuels for the transport sector: CO_2 captured from the atmosphere is combined with H_2 produced through renewable sources (wind, solar, nuclear) to synthesize clean liquid transportation fuels, such as diesel or gasoline, adding little or no carbon emissions to the atmosphere. A commercial validation project is starting in 2019, to test the integration risks at a larger scale.

Coaway

Coaway is a manufacturer proposing to use existing cooling towers of power plants to move large volume of air as an inlet for a carbon capture process. The idea is to surround the inlet of cooling towers with the absorption apparatus, so that large amounts of air can be processed quickly, bringing down costs. Similarly to CE, Coaway design captures CO_2 in a chemical reaction with an aqueous alkaline solution, involving potassium carbonate and bicarbonate $K_2CO_3/KHCO_3$ as intermediate species. The resulting material is regenerated in a thermal process that also releases the captured CO_2 as a concentrated stream ready for commercial use or further sequestration.
Soluble sorbents precipitate after reacting with CO_2 in flue gas: these precipitates are then decomposed using waste heat of power plat, available at around 95°C, instead of burning natural gas on purpose. This would allow them to have a very small capture price, claimed to be even lower than 20 $/ton$_{CO_2}$, that is mainly related to the investment expenses and not the fuel: lot of skepticism has been moved towards these estimates as they were not supported by real demonstration programs. Indeed, few details can be found about their plant design as no open pubications is available online and all information are kept reserved, besides the Earth Virgin Challenge [17].

2.4 Amine-Modified Solid Sorbents

Recent studies have focused on the potential of amine-based adsorbents for DAC application, due to their advantage in the regeneration step: indeed, regeneration can take place not only changing temperature (i.e. involving high quantity of heat or large temperature swings), but also pressure, humidity, or a combination of these methods [25, 38]. Moreover, lower temperatures are needed for regeneration, around 80-200°C, due to the less strong bound formed with carbon dioxide molecules.
Moreover, they show a fast kinetics of reaction and they are easy to prepare using inexpensive starting materials. Being solid, no separation or heating of water is required during the entire capture process.

This type of sorbents is already used at a commercial level to capture concentrated CO_2 in flue gases from power plants, for carbon dioxide removal from CO_2-rich natural gas streams, and in submarines to purify breathing air, with a potential for capturing the gas also from more dilute conditions as in the atmosphere.

By far, most of the reports on DAC applications have investigated the use of solid-supported amine materials, creating strong bonds with a good selectivity [32]: in this field a lot of work is still needed to identify the optimal match between the amine-based sorbent material and the solid support [39, 38].

Amine-functionalized adsorbent can be classified according to the interaction between the support and the active sorbent [40, 25]:

1. *Class 1: Physically Adsorbed Amines and Polyamines*, including monomeric/polymeric amines physically adsorbed on a supporting material, often silica. Sorbent is prepared by impregnating amines into the pores of the support. The physical interaction is weak, so that there is a degradation of amines and a reduction in adsorption performances due to amine leaching.

2. *Class 2: Chemically Immobilized Amines and Polyamines*, including monomeric/polymeric amines chemically bound to the support, so to reduce the issue of degradation with a permanent immobilization. The chemical reaction takes place between hydroxyl OH groups on the surface and the alkoxysilane 'anchoring' groups of the amines. Therefore, a covalent bond is formed to the solid substrate, usually a porous material: different types of surface are employed, from mesoporous silica to oxide, metal or polymer, provided that they have accessible hydroxyl groups.

3. *Class 3: Hyperbranched Aminosilicas*, including inorganic support and a chemically grafted polyamine covalently bonded to the solid support. Amine monomers are polymerized *in situ*, so to have polyamine structures tethered to the walls.
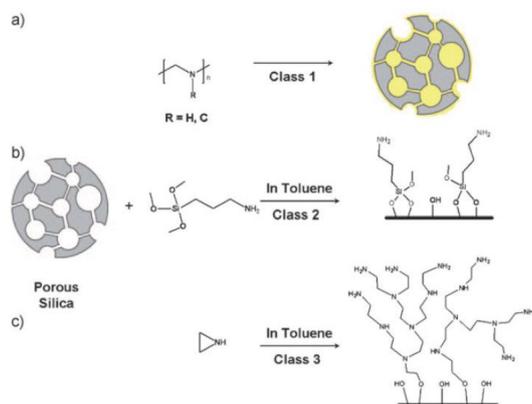

Figure 2.7: Different classes of amine-modified solid sorbents.

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2.4.1 Technical Design

The capture of CO_2 using amine-modified materials was first described by the company Global Thermostat (GT) in a 2007 patent application [41], applying sorbent materials referred to as hyperbranched aminosilicas, that pertains to Class 3 materials. Later on, in 2012, Global Thermostat patented a detailed air contactor capable of working with many sorbent media, including supported amines (Class 1). In both cases, a typical temperature swing adsorption (TSA) cycle was described, with desorption being achieved by an inert gas flow. Nevertheless, in a later patent by the same company, a steam flow was proposed as a substitute of inert gas to obtain more concentrated CO_2 in a more efficient way [32].

Class 1. Hybrid adsorbents:
Physically Adsorbed Amines and Polyamines
On the solid support, monomeric or polymeric amines are physically loaded through a simple preparation procedure. The most suitable amines for carbon dioxide adsorption, in terms of stability and CO_2 uptake, are silica-supported polyethylenimines (PEIs) [42], in particular the branched ones with both a low molecular weight ($M_w \sim 800$) and high molecular weight ($M_w \sim 25000$). The high sorption capacities of PEI[8] is related to the presence of highly accessible amino-groups at each chain end (as in can be seen in figure 2.8a), making them the benchmark adsorbents for CO_2 capture from air. This class of absorbents represents a promising option: they are quite inexpensive, easy to prepare, with significant CO_2 adsorption capacities, good kinetics under both dry and humid conditions and good regenerability, as either temperature and pressure swings with sweeping gas can be combined [42]. Moreover, humidity may also improve the adsorption of CO_2. Their main drawback is the loss of sorbent material and the low stability during regeneration, due to the lack of a strong chemical bond.

Silica materials are the most common supports for amine adsorbents, but some alternatives have been investigated, such as as alumina, and titania, mesoporous materials or carbon fibers. In particular, alumina supports are more resistant to structural changes and degradation under steam-stripping regeneration conditions compared to silica.

This cathegory is the one currently used in Global Thermostat demonstration plant (See following Section 2.4.2).

Class 2. Hybrid adsorbents:
Chemically Immobilized Amines and Polyamines
Some studies have been done on the removal of CO_2 from dry and humid air by adsorption using an amine-functionalized silica [43, 44]. After optimizing the experimental conditions, the CO_2 can be captured from an air flow even with high relative humidity (around 40%), with a good stability during regeneration. In this case the desorption took place with temperatures from 75 to 90°C.

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[8]It allows to take up 147 and 130 mg$_{CO_2}$/g, respectively, at 70°C from a pure stream of CO_2 at atmospheric pressure.

In particular, amine-functionalized nanofibrillated cellulose (as the ones shown in figure 2.8b) is the technology employed by the company Climeworks in his demonstration plant (See following Section 2.4.2).
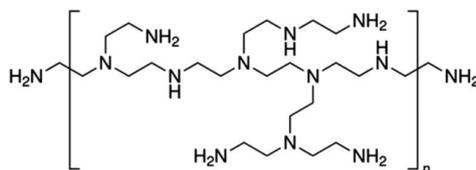
(a) Structure of a branched Poly-Ethylenimine (PEI) - Class 1
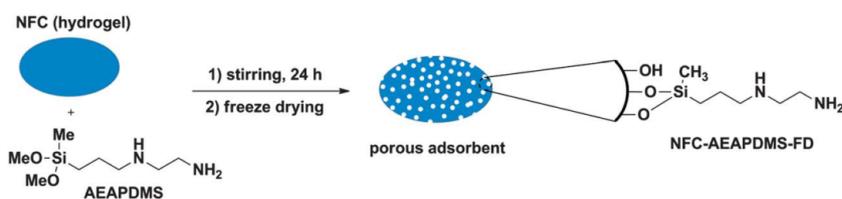
(b) Covalent immobilization of amine to a cellulose backbone - Class 2

Figure 2.8: Chemical structure for Class 1 and 2 amine-based adsorbent

Class 3. Hybrid adsorbents:
Inorganic Support and Grafting
This class tries to combine the advantages of chemically attached adsorbent materials of Class 2, in terms of low volatility and higher stability, with the high nitrogen loading of polymeric amines of Class 1. This can be obtained creating covalent bonds between the polymer backbone of the sorbent and the support, resulting in a robust and regenerable hybrid material for DAC applications.

Amine-modified adsorbents are considered one of the most promising option for DAC and future large scale applications from a twofold perspective: first, this plant design shows higher modularity than the one based on hydroxide solutions, as no big piece of equipment is needed (e.g. oxygen-fired kiln); secondly, the need for low-temperature heat during regeneration allows to integrate these capture facilities with waste heat coming from industrial processes and other power plants, reducing the overall capture cost. Nevertheless, few technical details about plant layout are available in the literature, as they are being implemented by innovative companies, Global Thermostat and Climeworks, aiming to keep their competitive advantage on the market.

2.4.2 Existing Companies and Demonstration Plants

Global Thermostat

Due to their promising potential, amine adsorbents have been the focus of recent private research on DAC, but little progress has been proven in the field. The start-up Global Thermostat (GT) was founded by Peter Eisenberger, a professor from the Columbia University, and employs an amine-based chemical sorbents bonded to a

porous honeycomb ceramic monoliths (Class 1 material, as discussed previously).
GT is located at the Stanford Research Institute (SRI) in Silicon Valley since 2010, where they built the first demonstration plant for its amine-based process in 2015, comprising 640 ceramic cubes embedded with the amine sorbent. The air and/or the flue gas mixture are moved by fans over a wall of Corning's honeycomb monoliths, which are coated with the proprietary sorbent. The coated monoliths adsorb the CO_2, then process steam is used to desorb it from the wall, obtaining high purity CO_2.
One of the main advantage is the modularity: the single unit has a capacity of 50 000 ton/yr and could be scaled up to a 40-modules power plant able to capture 2 Mton/yr. Since the units can be stacked, the footprint is also reduced.

Figure 2.9: GT demonstration plant at Menlo Park, California, operated since 2010

Two contactors are alternated in a batch process: one collects CO_2 from ambient air while the other is being stripped with steam at 85°C, at a temperature even lower than the amine-based capture used for CCS from power plants [9]. The system will use low-temperature steam for a twofold purpose: it heats the absorbent surface, releasing the CO_2 to be collected, while blowing CO_2 away from the surface. As a consequence, the heat-management part of the system results much simplified, as well as the design of the scrubber, reducing the overall costs. Moreover, the need for low-grade steam allows to combine the capture of CO_2 with heavy industrial processes (such as metals melting, cement production, and petrochemical refining) that provide waste heat.

Eisenberger estimates low energy and economic costs of large-scale implementations, with an overall capture price about 50 \$/ton$_{CO_2}$, though these estimates are affected by uncertainties about the lifespan of the amine-base adsorbents used [45]. Volatility, degradation of performances of the amine and/or its support material, and the cost of production are all critical factors that need to be further studied with the specific framing of practical DAC application, as they could increase the overall capture price with respect to the optimistic estimates: as all demonstration plants are being operated for few years, the actual duration of these sorbents cannot be yet evaluated. They claim that their system does not require any government subsidy to be economical and profitable, as CO_2 can be used in other existing markets, such as food and beverages, plastics, greenhouses, synthetic fuels and industrial applications. In order to

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[9]Note that post-combustion capture with MEA from coal and gas-based power plants is applied to flue gas at around 70°C, while amine regeneration takes place at 120°C

assess the scalability of this integration, the start-up has a partnership with a Nevada-based company called Algae Systems to make biofuels using carbon dioxide and algae. Another interesting option would be to combine CO_2 capture from air with capture from the flue gas of a power plant: using the power plant's low cost process heat to provide the energy needed for the air capture process, GT technology may eventually transform power plants into net carbon sinks, capturing at the same time CO_2 both from the power plant emissions and from the air. Global Thermostat technology can also work with renewable power plants, such as concentrated solar and nuclear plant, because it captures carbon directly from air using the plant's process heat.

Climeworks

Class 2 of amine-based adsorbents has been used for the first time in a cyclic temperature-vacuum swing (TVS) process by Wurzbacher in 2011 [44], and this is the technology on which the company Climeworks is building its business.

This company is a spin-off from ETH Zurich, founded by engineers Christoph Gebald and Jan Wurzbacher, that started researching on DAC applications during their master studies, developing the first prototype in 2009.
All plants developed by Climeworks, which are already commercially available, are modular and scalable: the basic unit consists of one single collector, with a shipping container size, which contains six filters, being able to capture 135 kg CO_2/day (around 50 ton/year), with a footprint of 20 m^2. Climeworks machine consists of a series of three, stacked units; a large hot water storage tank sits alongside, together with two further containers housing control equipment.
In October 2015 they sold the first commercial direct CO_2 capture plant at Hinwil, a small town just outside Zurich, composed of 18 single units, with an overall capacity of 900 ton_{CO_2}/yr: the plant is sited in a favourable location, on the roof of a municipal waste incinerator, which supplies the low-grade heat that it needs, below 110°C [46].

Carbon dioxide is captured through an amine-functionalized cellulose filter, made of porous granulates, which turn to be particularly suitable for DAC as amines react selectively with carbon dioxide in the atmosphere, even in presence of moisture, at ambient temperature and pressure. Then high purity CO_2 can be desorbed with a one-step temperature-vacuum swing. Up to now, the filter has been proved to last several thousand cycles, but it has being in operation only for a limited time span.
Total thermal energy requirements are between 5.4 and 7.2 GJ per ton of CO_2 captured, requiring hot water at around 100°C for regeneration and cooling water at temperature lower than 15°C. In addition to that, electricity needs are stated to be around 10% of the total energy required.
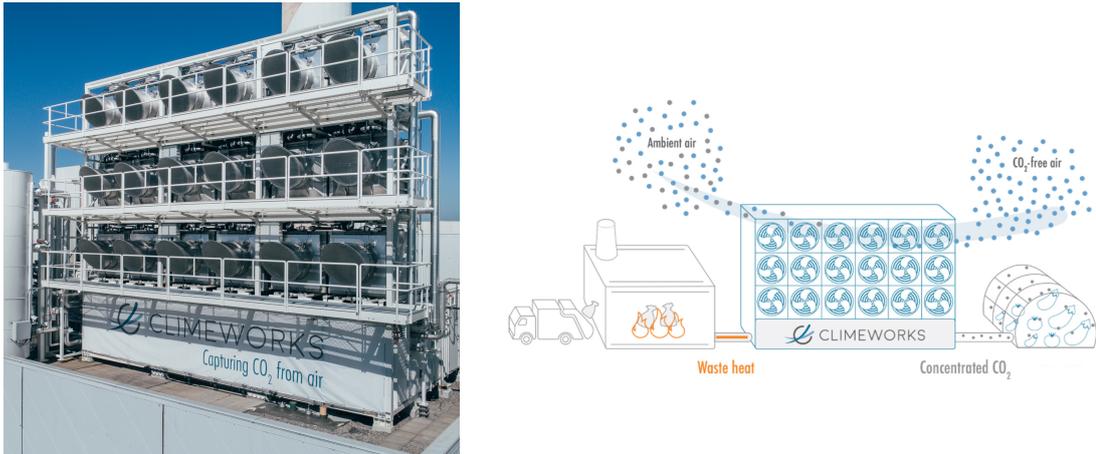
Figure 2.10: Climeworks Plant Setup CitewebClimeworks

The concentrated gas stream collected is then supplied to customers or sent to sequestration sites, addressing the same sectors as Global Thermostat: currently, the demonstration plant is selling the captured CO_2 to local greenhouses, to boost the growth of vegetables, but it is worth noting that this is not covering its full costs [47]. In the past two years, Climeworks has grown rapidly, reaching 45 employees today: its 20m\$ in financing includes 5m\$ in Swiss government grants and 15m\$ from private equity. In cooperation with the Danish company Union Engineering they have also started developing a plant supplying CO_2 for beverages. Their ultimate goal is to capture 225 millions ton of CO_2 in 2025, that is equal to around 1% of global emissions. Note that this is currently not possible only by commercial means, as it is not a competitive option yet and the costs cannot be covered by selling out the captured CO_2 to existing industries. Therefore, a strong political will is required to support such technologies, for instance with a price on carbon. Moreover, also technical constraints need to be considered, as to achieve this ambitious goal 750 000 units would be needed, while the current production line of Climeworks has an annual capacity of 100 units.

2.5 Other Solutions

Besides amine-based ones, other inorganic solid sorbents have been researched, and some of them have already been applied to capture CO_2 from air, for submarines or space cabin application and air purification: on the one hand, ion-exchange resins, known also as *Artificial Tree*, are being developed by Lackner, gaining a good level of attention in the scientific debate, while new innovative materials are being investigated, such as Zeolites, Metal Organic Frameworks (MOFs) [32] or Alumina [33] and solid oxide [34].

The work is still focused on identifying the most promising materials and consensus has not been reached yet on the most favorable category. On the one hand, zeolites may need far lower temperatures (around 240°C) for regeneration than other materials, but need also pressure changes to operate, while showing extreme sensitivity to atmosphere

humidity [48], thus requiring air to be dried before contacting. On the other hand, solid Na- and Ca- oxides have an operating principle very similar to their liquid counterpart (see section 2.3), but suffer from slow CO_2 uptake and a high sensitivity to humidity as well. As the air need to be dried and heat needs to be provided during CO_2 uptake, this increases the energy requirements significantly. According to first studies [34], the thermal energy needed is about 241 GJ/ton of CO_2 captured, far higher than other systems suggested, with additional sorbent degradation issues, especially for CaO systems.

2.5.1 Artificial Tree

The focus of C. Lackner research at the Center for Negative Emissions (Arizona) has been on developing an appropriate sorbent for DAC applications since 2009, starting with strong aqueous alkaline solutions [31], then moving to solid sorbents, due to the high energy requirements and the kinetic limitations of hydroxide chemistry. In 2010, Lackner and Global Research Technologies LLC described in a patent application [49] the use of anionic exchange resins for DAC, where quaternary amines are attached to a polystyrene backbone. It is referred to as "artificial trees", as it mimics the process of capturing CO_2 from the ambient that regular trees do.

Each amine group carries a permanent positive charge: it is similar to an ammonium ion NH_4^+ , where an organic carbon chain attached to the polymer matrix replaces each hydrogen. The resin acts as a strong base, as the positive ions anchored to the polymer backbone never releases a proton and never comes back to the analog of NH_3, combining the low binding energy of carbonate to bicarbonate with a faster reaction kinetics [31]. It is a composite material with a resin similar to Marathon A (provided by Dow Chemicals) as the active ingredient: it is produced as an electro-chemical membrane by Snowpure LLC (San Clemente, California), where small resin particles are embedded into an inert polypropylene sheet[10].

The maximum theoretical carbon loading of this resin is defined by the total number of positive ionic charges available on the resin, and it is comparable with the uptake of strong base solutions. Moreover, the exchange resin are capable of adsorbing CO_2 when it is dry and releases it when wet (i.e. humidity or moisture swing), obtaining a CO_2 enriched stream of air of up to 5% or 50 000 ppm. It should be noted that the stream released is characterized by a low purity, but it still represents an increased concentration compared to the starting point at 400 ppm. The output dilution results similar to flue gas, but located at the site where it is needed and without the presence of contaminants.
In order to obtain high purity stream of CO_2 to be sent to sequestration sites, a second step is required, similar to CCS technologies applied to power plants, thus increasing the overall capture cost. Alternatively, the low purity stream can be used directly in greenhouses to enhance plants and algae growth [50].

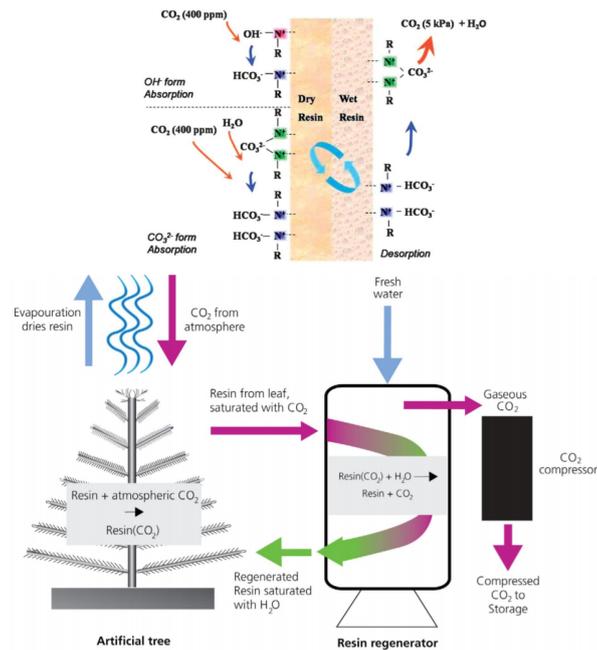[10]The resin makes up about 60% of the weight of the material

Figure 2.11: Ionic exchange membrane: working principle, according to Lackner researches [31].

The main advantage of humidity swing technologies is that moving large volume of air does not involve additional cost as it relies on ambient wind: as wind conditions are used both to drive air through filters and later to dry them for regeneration, the energy consumption of this capture process results relatively low. The drawback is that performances are highly weather and geography dependent. This dry-wet reaction cycle is more similar to the removal of trace amount of contaminants rather than traditional capture from flue gases, and can be adapted for continuous operation, not showing degradation after many regeneration cycles.

Process Steps

The modular design include a set of filters (2.5 m tall, 1 m wide and 30 to 40 cm thick), with a flow speed through them around 1 m/s: a compact container-size device will be composed by 30 of these single, capturing 1 ton/day. The first prototype was actually built by the Center for Negative Emission, but no scientific publications based on its performances have been issued yet. Five main steps are involved in the design proposed by Lackner [31, 51]:

Step 1: CO_2 Desorption, Moisture Swing
Each unit consists of six regeneration chambers, each holding five air filters: chambers are arranged in a circle, with one interacting with atmosphere while the others are going through different stages of regeneration to extract CO_2 from the resin.
Driven by ambient wind, air flows through the filters where the resin adsorbs some carbon dioxide, reducing CO_2 concentration in the air from 400 ppm to around 360 ppm: once saturated, the filter is removed from the air collector and moved on an

automated conveyor system to be regenerated. When the chamber is filled with saturated filters, air is pulled out creating vacuum (0.01 bar) and moisture is injected, either by exposing the inside of the chamber to a reservoir of warm briny water, or by spraying clean water into the chamber, so to release CO_2. The water vapor stream will move in a counter-flow manner from chamber to chamber, gradually increasing the partial pressure of CO_2, and will be eventually pumped out of the last chamber with a CO_2 partial pressure will be between 5 and 10 kPA, thus requiring to be further compressed up to pipeline pressure in the next steps.

The energy required for sorbent desorption consists mainly of electricity to remove ambient air from the desorption chambers: this step requires around 4 kJ/mol of CO_2, representing only the 10% of the total energy consumption.

Step 2: CO_2 Drying

During desorption, the sorbent gives off a mixed stream of CO_2 and H_2O, therefore in the first compression stages, a big amount of water will condense. The removal of water is achieved by condensation via a cold trap cooled by a heat pump, releasing the associated condensation heat (around 40 kJ/mol_{H_2O} that correspond to 23 kJ/mol CO_2): this heat is used to mantain the temperature of regeneration chambers above the ambient one. In this way the regeneration system only requires mechanical energy, and the heat needed will be produced internally as a by-product of compression.

Step 3: CO2 Compression

CO_2 needs to be compressed from a partial pressure of 5 kPa to 6.7 MPa[11]. This is the most energy-intense step, requiring 19 kJ/mol CO_2 of electricity to drive a compressor. As for the previous drying step, different portions of CO_2 desorb at different pressures, determining the pressure of each desorption chamber: therefore, each portion requires different compression work.

Step 4: Sorbent Regeneration

During regeneration the sorbent is returned to its thermodynamic base state, mostly dry and with low CO_2 loading. The required energy for this step comes predominantly by ambient heat, provided by air with 30% humidity and 20°C temperature, that causes the sorbent to dry.

Step 5: Auxiliary Processes

The steps above cover all energy-intensive processes of the proposed DAC system. Additional electricity is needed to move sorbent embedded in filters in and out of the desorption chamber, through an horizontal conveyor-belt structure (0.7 kWh/ton), and to compress water from 1 to 3 bar with a pump to be sprayed on the filters in the evacuated regenerator(1.3 kWh/ton). Furthermore, power electronic controls, sensor, and actuators for water and air valves require 11 kWh/ton.

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[11]Note that at this pressure, with $T = 300K$, carbon dioxide turns liquid, therefore reducing the energy needed for further compression

Considering inefficiencies of pumps/compressors and the mechanical operation of the air collector, the overall energy consumption is 50 kJ/mol$_{CO_2}$, that is 1.1 GJ/ton$_{CO_2}$.

Existing Companies

Based on the research by C.Lackner, the Center for Negative Emissions has been improving the anionic exchange resin and a startup was founded to commercialize this system, called **Kilimanjaro Energy**. Unfortunately, this closed recently due to a lack of fundings. According to Lackner's vision, ten million 'artificial trees' could remove 3.6 billion tonnes of carbon dioxide each year (3.6 Gt/yr), causing CO_2 atmospheric concentrations to drop by about 0.5 parts per million per year.

Through Infinitree LLC, a corporate venture, the technology developer **Carbon Sink** can capture carbon dioxide from air, using a system which is an evolution of the one developed by Kilimanjaro Energy (i.e. Lackner prototype). The difference is in the application: in order to be economically sustainable as a company, the focus of Carbon Sink is to target sustainable low carbon opportunities for using the CO_2 captured from the atmosphere today. As long as for now, the system proposed by Infinitree is coupled with greenhouses, as CO_2 allows to enhance the growth of food with less water and fertilizers. Future proposed applications include the production of carbon-neutral biofuels and clean water, enhancing the growth of aquatic plant chambers to purify it.

2.5.2 Solid Inorganic Sorbents: K_2CO_3/Alumina Composite

Potassium carbonate K_2CO_3 has been initially proposed in aqueous solution (similar process to the one for NaOH or $Ca(OH)_2$): the problem of low reaction rate encountered can be overcome using a composite material that has K_2CO_3 dispersed inside a porous matrix. In particular, mesoporous $\gamma - Al_2O_3$ has been proposed, on which K_2CO_3 particles can be dispersed, as this class of material has already been used for traditional CCS from flue gas. The composite sorbent with $K_2CO_3/\gamma - Al_2O_3$ is obtained by drenching the pores with an aqueous solution of potassium carbonate and then drying it.

According to Veselovskaya research [33], the absorption capacity of the material increases significantly decreasing the grain size, which indicates that the process is limited by mass transfer. Increasing the regeneration temperature (from 250 to 300°C), also CO_2 absorption capacity is enhanced. Absorption performances show good stability, therefore it could be considered as a promising material for direct capture from air.

Veselovskaya studies focused on the integration of DAC plants to produce synthetic and renewable methane as a way to store hydrogen, so to provide a valuable feedstock for the power-to-fuel industry. As a concentrated stream of CO_2 can be obtained at the outlet, it can be used directly in subsequent processes as a reactant for methane synthesis (i.e. *methanation*): in this case, the heat generated during the reaction can be used for the thermal regeneration of the sorbent, with an overall efficiency around 50%.

This material is still at the research stage, with no company having already implemented it in a demonstration plant. On a laboratory scale, Veselovskaya tested its capture performances in a cyclic test apparatus, with 4 absorber connected in parallel so that to test different materials at the same time. The cycle of Temperature-swing Adsorption (TSA) has two steps: absorption and regeneration, each with a time span of 6 hours. It has been demonstrated that the material is still thermally stable after multiples temperature-swing cycles not suffering degradation in oxidating environment, as amine-based ones do. At the same time it isnot influenced by the presence of moisture, differently from zeolites that show high affinity with water and humidity.

2.5.3   Ca- and Na-based Solid Sorbents

The main problem of carbonation of solid sodium oxides at ambient temperature is the slow reaction rate: as a consequence a large mass flow rate is required and the overall process turns to be not feasible from a technical and economic point of view.
Ca-based cycles have more favorable kinetics than Na-based, as the carbonation of CaO and $Ca(OH)_2$ into $CaCO_3$ is faster and can be catalyzed by the presence of water in the stream. Unfortunately, the biggest drawback is that the reaction temperature for the carbonation process is around is 300-400°C, much higher than for sodium oxides. To solve this issue, the proposal was to use Concentrated Solar Power as a heat source [34], to sustain both carbonation (at 375°C) and the subsequent calcination of $CaCO_3$ (at 875°C). Overall the energy required is about 10.6 MJ/mol, that is 241 GJ/ton: being one order of magnitude higher that all other technologies and requiring to heat a large volume of air to high temperatures for carbonation, this solution does not appear to be feasible on a commercial scale.

2.6   Technology Summary

After having analyzed the different technology alternatives available, the main characteristics of the various categories can be summed up as follows.

The thermal energy consumption of $CaO/CaCO_3$ carbonation is pretty high[12], so that it is not a feasible option on its own. Therefore, coupling NaOH and KOH solution with lime carbonation is the base for DAC processes based on **strong base solutions**: energy consumption can be reduced, while equipment are already available and widely used in other industrial processes. The main disadvantage for this process category is the high temperature required for regeneration, around 800°C, and the need to use an oxygen-fired kiln to avoid further emissions.

On the other hand, DAC systems based on **amine-functionalized adsorbents** does not require any conditioning and regeneration can be performed with low-grade heat at around 90°C, which is commonly available as industrial waste heat, as well as solar or geothermal heat. The possibility to use waste-heat to fulfill thermal energy

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[12]It is around 2500 kJ/mol, that is almost 55 GJ/ton

needs will reduce operational costs for this DAC technology, while ensuring high purity stream of CO_2.

Using an **ion exchange resin** does not require any thermal energy for regeneration as it is based on humidity swings: as a consequence, the capture of CO_2 is limited to dry air conditions (below 40%) and it results in a stream with low purity, so that a further concentration step is needed to store it. This means adding a CCS unit to the overall plant before sending carbon dioxide to storage sites, increasing the cost for this capture process.
Other solid adsorbents, such as zeolites and solid CaO-based ones, require energy inputs still too high, with lots of uncertainties on their actual performances.

In conclusion, DAC systems featuring NaOH and amine-functionalized adsorbents are generally considered the most technically feasible. The economic viability of such systems can only be judged once detailed engineering review of demonstration plants will be available. Up to now, few specific data can be found in the literature, but an exhaustive assessment has been published by D.W Keith and G.Holmes [35] while I was working on my thesis[13], based on the performances of Carbon Engineering plant in Canada, and further analysis are expected to come out in next months as this technology is drawing more and more attention in the scientific community.

Sorbent	Regeneration	Advantage	Disadvantage
Strong Base $Ca(OH)_2$/NaOH/KOH	T > 800°C	low cost sorbent simple sorbent handling	high thermal energy need
Amine-functionalized adsorbent	T ~100-120 °C	low regeneration temp high purity CO_2	Complex sorbent handling Sorbent degradation
Ion Exchange Membrane	T ~90-100 humidity swing	no thermal energy for regeneration	Low purity CO_2 Complex sorbent handling Dry climate needed

Table 2.1: Summary of available DAC technologies

2.7   Energy Estimates

Estimates about the energy required by DAC processes are largely discussed: while the theoretical requirements for separating carbon dioxide from ambient air are quite small, in practice systems need higher energy input, as already discussed in section 2.2.2. Moreover, it needs to be considered that different technologies require different amount and different type of energy.

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[13]Published in June 2018 on the journal Joule

According to the review done by the Postdam Institute for Climate Impact Research [27], thermal energy needs provided in the relevant literature range between 6 to 10 GJ/ton, while electrical ones are between 1.1 and 1.9 GJ/ton of CO_2 captured.

Considering individual technology options, **aqueous solutions of strong base** require between 6 and 8 GJ/ton of high-temperature heat, provided by burning natural gas, and 1.6-1.8 GJ/ton of electricity, according to the APS report. When applying KOH instead of sodium hydroxide NaOH as CE is researching, these energy needs can be lowered to about 1.3 GJ/ton of electricity and 5.3 GJ/ton of heat.

As it has been already highlighted, the used of **amine-based solid sorbents** allow to further reduce the energy required in the regeneration step. In particular, Climeworks estimates are between 5.4 and 7.2 GJ/ton of low-temperature heat and 0.7 to 1.1 GJ/ton of electricity. The values provided by Global Thermostat are similar, with thermal input between 4.3 and 5.2 GJ/ton and electricity input around 0.6 GJ/ton.

Differently, the **artificial tree** proposed by Lackner has the peculiarity of requiring only electricity to be operated, as the regeneration takes place through a humidity swing process, thus not involving any change in temperature. He claims that this technology only requires 1.14 GJ/ton_{CO_2} of electricity , making it an interesting option compared to other processes. It should be noted that additional electricity will be required to have a stream of high-purity CO_2, with consumption similar to the capture process applied to flue gases in post-combustion CCS.

2.8   Cost Estimates

Currently, estimates about capital and operating costs for DAC Plants are quite uncertain, ranging from 30 \$ to 1000 $\$/ton_{CO_2}$ [52]. When referring to DAC plants, the scientific community uses APS evaluation around 600 \$/ton CO_2 as the main benchmark, even if it does refer to a specific technology option, namely the use of strong bases. An exhaustive investigation of different estimates across the literature and the ones claimed by companies has been performed for my research work, so to have a clear picture of possible cost reduction and targets for the future. It should be noted that detailed cost assessments are available only for technologies based on hydroxide solutions, with a breakdown down to the individual component level, while very few data can be found in the literature about plants with amine-based sorbents and ionic membrane, as information are not revealed by the companies involved.

1. Aqueous Solutions of Strong Base

For this group of technologies the main cost reference is represented by the APS report of 2011 [20]: the overall price for capturing carbon dioxide from the air is about 430 \$/ton of CO_2 captured (that is 610 \$/ton of CO_2 avoided[14]), including energy costs

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[14]Note that the difference between cost per ton of CO_2 captured and ton of CO_2 avoided depends on the carbon intensity of the fuel used to supply the heat and the electricity needed to the plant. As in the model used for my thesis work, these additional emissions are accounted endogenously by the

for both electricity and fuel. According to the design optimization of the air contactor provided by Mazzotti [29], the cost can be reduced down to 376 $/ton of CO_2 captured.

It should be noticed that both estimates refer to a plant setup based on existing technologies and not properly designed for air capture, therefore they should be considered as the high end estimation. A low cost scenario comes from Holmes and Keith research([30]): as it has been already highlighted, they proposed a completely different design for the contactor, based on the slab concept developed by CE, bringing its cost down to about 60 $/ton CO_2, that is four times lower that APS estimate. Overall, capture cost results between 280 and 350 $/ton of CO_2 captured, in the optimistic and pessimistic case respectively[15].
Traditional packed towers, as the ones considered by APS, imply lower technical risk as they are already used for CO_2 stripping applications, but lead to really high cost.

Recently, updated assessment on hydroxide-based plants have been provided, building on on the operations of the demonstration plant by Carbon Engineering [35]: the initial cost estimated is about 230$/ton of CO_2 captured, including energy needs. Considering learning effects and improvements in capital and construction costs indicated by vendor and engineering firms, potential cost reduction brings it down to 170 $/ton, when scaling the market size. The floor cost provided is about 150 $/ton, referring to a plant configuration with minimum gas input and most of the energy needs supplied through electricity: this alternative may apply in case of high abundance of carbon-free and low-cost power, that should be the main paradigm in a future low-carbon world.
Estimates as small as Coaway plants (20 $/ton) seems to be too optimistic and not supported by a rigid techno-economic analysis, neither by the construction of real demonstration plants.

		APS, 2011	Mazzotti, 2013	Holmes, 2012	Keith, 2018
CAPEX	[M$]	2200-2900	1930-2200	1220-1650	700-1150
	[$/ton]	260-350	220-260	150-200	85-140
OPEX labor, maintenance	[$/ton]	90-120	76-90	50-70	30-40
OPEX with energy	[$/ton]	170-200	156-170	130-150	60-80
TOT	[$/ton$_{captured}$]	430-550	376-430	280-350	150-230

Table 2.2: Capital and Operational Cost estimates for plant using aqueous solutions of strong bases, according to the available literature [20, 29, 37, 35].

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model, we are simply interested in the cost for capturing 1 ton of CO_2 with DAC plants.

[15]Note that also a study carried out by Zeman [53] provides a similar cost estimate around 300 $/ton.

2. Amine Modified Solid Sorbent

For this category, very few details on cost assessments can be found in the literature, as it has been mostly developed by private companies, keen on keeping information secret to protect their competitive advantage. Therefore we will refer mainly to the estimates provided by these companies about future cost targets, even if these figures must be considered carefully, as they are likely to be excessively optimistic.

During conferences and public events to promote their technology, *Climeworks* was able to provide solid cost estimates, starting from 600 $/ton$_{CO_2}$ today for their first demonstration plant (partly covered by selling the CO_2 to a nearby fruit and vegetable grower for use in its greenhouse), with a rapid cost reduction in next years. Indeed, they foresee a threefold cost reduction in next 5 years, so to have a capture cost around 200 $/ton, with a long term target price of 100 $/ton by 2030 [47]. In order to achieve it quickly, economies of scale will play an important role, that means building more plants and purchasing larger volumes of material, as well as automating the production steps. To further reduce the capture cost below 200$/ton, fundamental R&D will be needed to find cheaper materials, as simply scaling the market won't be enough. The main unknown is still related to the lifetime and performances of amines in time, as this will impact significantly on the overall capture cost.

Considering *Global Thermostat*, a significant lower price for their prototype is claimed, ranging between 15 and 50 $/ton of CO_2 captured, depending on the lifetime of amines. Such small cost can be achieved as the steam needed for the process come from waste heat or process heat at low or even no cost. The advantage is also that only steam and electricity are consumed, potentially with no additional emissions.

3. Artificial Tree

When dealing with the technology developed by Lackner, few details on how capture cost is computed can be found in the literature and this represents one of the main criticism that has been moved to it. Indeed, no demonstration plant has been built yet to support the optimistic estimates, which appear to be much less rigorous than the ones presented in APS report. According to [31], the first prototype is expected to have a cost around 200 $/ton CO_2, going down in the future to 30 $/ton[16], due to learning by doing and further improvements, mainly related to the development of the material, so to reduce the amount of resin needed and therefore the size of the system. As the main cost components are the resin itself and the regeneration chambers, accounting together for almost 70 % of the total cost, these improvements may reduce a lot the overall capital cost of about 90%, going from 200 000 $ to 20 000 $ for each unit. Decreasing the amount of resin in the future, the lower bound for cost reduction is represented by the cost of electricity and pumps/compressors.

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[16]It should be noted that this price includes also electricity cost, which is around 15$/ton CO_2, Considering a price of electricity of 5 cents/kWh.

For other solid sorbents no cost estimates can be found in the literature as they are still researched at a lab scale, with no plant design being proposed yet.

2.8.1 Cost Targets and Niche Markets for CO_2

At this point, the technological immaturity of DAC technologies means that any kind of estimates about future costs, performances and scalability are merely speculative, as it is difficult to account for learning mechanisms achieved when building the first systems. Indeed, the costs of new technologies can drop by orders of magnitude as they develop and mass production follows: for instance, the cost of solar panels has dropped almost 100-fold since the 1950s. Moreover, policy support can also make the difference: once sulfur emission trading was made into law in the 90s in USA, sulfur reductions at power plants proved to be 10 times cheaper in a 4 year period than experts predicted shortly before the start of trading.

Therefore, there is a urgent need for further assessment about DAC potential given different scenarios for cost reduction and energy performance improvements, as it will be discussed later in reference to my reasearch work in Chapter 4: in this perspective, IAMs can represent a useful tool to perform a range of sensitivity analysis.

Making Direct Air Capture a useful technology depends also on the market value for CO_2 as a chemical commodity [24]: Climeworks and its competitors are showing that, if DAC can be made cheap enough for being of commercial interest, then the stream of captured CO_2 can be used as a valuable feedstock for various applications, such as the synthesis of fuels, the production of synthetic intermediates for pharmaceuticals or other chemical products, as well as for feeding greenhouses and algae cultivation. In a free market the removal of CO_2 from the air starts to make economic sense from a production price around 100\$/ton, as this is the price that oil companies would pay for liquefied CO_2 to be injected into reservoirs to squeeze out oil (i.e. Enhanced Oil Recovery): starting from the baseline of 600 \$/ton [20], the challenge is yet large, but comparable to corresponding cost reduction required for other climate mitigation technologies. Bringing down cost, air capture technologies could serve other markets beyond the oil industry.

It should be noted that all these proposed applications do not have a size large enough to really have a climate impact if DAC is expected to be developed massively as a NET option[17]. Nevertheless, they could represent an incentive to develop DAC technology in the near term, with geological sequestration being still the ultimate goal as a long-term mitigation strategy [32].

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[17]As a comparison, climate mitigation will require negative emissions between 10 and 35 Gt_{CO_2}/yr, while the Global CCS Institute reports that the cumulative global demand for EOR may be around 500 Mton in 2020 [54]

Chapter 3

Methods

Engineering models represent a useful tool to understand a complex problem and its potential solutions, considering a range of different implications and effects, before undertaking the expenses and efforts of a full implementation. Integrated Assessment Models (IAMs) have been developed to represent the complex interactions between climate science, economics and the energy systems, so to assess the feasibility of a range of mitigation pathways and inform policy-maker about the urgency of policy implementation and government support.

In Section 3.1 a general introduction on IAMs will be provided, before focusing on the specific model TIAM used in this work to assess the impact of Direct Air Capture deployment, in Section 3.2. Both the economic rationale and the representation of the energy system will be described, while the implementation of DAC technologies within TIAM will be discussed in the following chapter.
As additional tool, an Expert Elicitation has been carried out to better understand the potential of this new technology in the future from the perspective of specialists currently researching in this field, given the fragmentary literature available on this topic. The insights gathered from it are presented in Section 3.3.

3.1 IAMs: Integrated Assessment Models

Climate change is a very complex challenge that requires to understand the interactions between different fields of knowledge like climate, energy and economy. Carbon fluxes between natural systems (i.e. atmosphere, land, ocean) and commercial ones (i.e. electricity and heat production, transportation, industry) are highly coupled and introduce feedbacks in the overall global system, that makes prediction on overall behavior very difficult [55]. Therefore, a variety of system models need to be created to guide holistic policies for carbon management, developing approaches to systematically study parametric sensitivity and quantify uncertainties related to different variables.
It is important to understand that the main goal of simulations done with these models is to explore *what if* scenarios, so to identify performance and cost targets that would define a research, development and demonstration strategy for these technological

pathways: in this perspective, modeling exercises are a useful policy tool, even if they rely on a large number of assumptions, and need to be intensified.

IAMs or Integrated Assessment Models try to integrate a description of GHG emissions and their impact on temperature (*climate science model*) with a description of how climate changes may affect output, consumption, and other economic variables (*economic model*), to understand the basic mechanisms involved and the dynamics among key variables. IAMs generally assume that some sort of idealized equilibrium for the economy, or at least for certain energy technology markets, will be achieved in each year for which results are computed. The first model, based on a simple linear programming of supply and demand, was developed by W.Nordhaus back in 1977 [56]. During the last decade, these long-term scenarios have played a high profile role in the most important analyses of climate change, such as the Stern Report [57] and the IPCC's Assessments [2].
There are mainly two categories of IAMs:

- *Simulation Models*, in which equilibria are developed per each point in time. GCAM and IMAGE pertain to this category.

- *Optimization Models*, in which temporal dynamics is resolved at once, assuming a forward-looking planner with perfect foresight that discount future well-being. The model used for this thesis work, TIAM, is included in this class.

IAMs analyses over the long run are valuable as model internal consistency allows one to assess the relative implication of policy alternatives, and can help in estimating which policy strategy are likely to have higher or lower costs [58]. Indeed, unlike forecasts, scenarios do not assume perfect knowledge of the main drivers of the energy system: a scenario consists of a set of coherent assumptions about future trajectories of these drivers, leading to a coherent organization of the system under research.
They shouldn't be considered as a tool to make realistic mitigation plans decade by decade in the short term, as they lack of sectoral details and specialization. Nevertheless, trying to model long-term impacts and possible trends can be helpful in making strategic investment and decision in the short-term.

3.1.1 Criticism to IAMs

Generally, many criticisms have been moved to IAMs from a twofold perspective: on the one hand, the structure itself of these models is addressed, in terms of the social welfare utility function used and the functional form applied to determine the response of temperature to CO_2 concentration [59]. On the other hand, the inherent uncertainty related to any long term estimate is being criticized, as it would make their results useless from a policy perspective [60]. Moreover, it is usually pointed out that models ignore the possibility of a catastrophic event or climate outcome.

Leaving aside this third point, I personally think that the first two aspects represent a limitations of IAMs modeling approach researcher and policy makers should be aware of when presenting their results, nevertheless they are not a reason for disregarding IAMs as part of a portfolio of tools needed to tackle the climate change challenge. The modeler has a great deal of freedom in choosing functional forms, parameter values, and other inputs, and different choices can give widely different estimates for the social cost of carbon and the optimal amount of abatement. As a way to reduce the uncertainty inherent in any long-term forecast and the arbitrariness often criticized to IAMs, sensitivity analysis on key parameters is required, to check robustness of the model results. This aspect will be extensively considered in Section 4.6.

3.2   TIAM

The scenario outputs of this work have been produced with the TIMES Integrated Assessment Model (TIAM) owned by the Grantham Institute within Imperial College London. TIAM-Grantham is a multi-region, least-cost optimization model, minimizing the total present value cost of the global energy system (using by default a 5% time discount rate) to meet future energy service demands.

As already stated, energy system models inform policymakers about the potential importance of particular energy technologies by examining whether their presence or absence, with given costs and performances, has an impact on the overall costs of decarbonisation. Technology-rich models like TIAM (TIMES Integrated Assessment Model) and MARKAL (MARKet ALLocation) are particularly of interest in this perspective, to explore the least-cost evolution pathways of the energy system required to meet prescribed climate targets. TIMES is a model generator for local, national or multi-regional energy systems, developed as a successor of the MARKAL [61] and EFOM [62] bottom-up energy models, and incorporating the main features of these ancestors. This model aims to supply energy services at minimum global cost, with the objective function maximizing the net social surplus, when all markets are in equilibrium. It is a linear programming bottom-up energy model, that could be coupled with a climate module: demands for different energy services represent the main exogenous driver. Demands for energy services can be elastic to their own prices, thus capturing main feedback from the economy to the energy system [63].

ETSAP-TIAM is the global multiregional incarnation of the TIMES model generator, with a number of sets and processes already defined and built in it. TIMES represents the model mathematical structure, which is fixed, while TIAM represents the model instance which is generated by TIMES based on the input information provided by the modeler: these are based on the International Energy Agency (IEA) databases and incorporated in TIMES structure within the Energy Technology Systems Analysis Program (ETSAP).

TIAM can be defined as a *technology explicit, multi-regional, partial equilibrium model*, that assumes price elastic demands, competitive markets, and perfect foresight, resulting in marginal value pricing.

Technology Explicit Model: each technology is described in TIMES by a number of technical and economic parameters. It is also technology-rich model, so that technologies can be modeled purely via data input specification, without having to modify model's equations. This makes the model *data driven.*

Multi-Regional: TIMES models covering the entire energy system include up to 15 regional modules, while some existing sectoral TIMES models may consist of up to 30 regions. The number of regions in a model is limited only by the difficulty of solving linear programming of very large size. Individual regional modules are linked by energy and material trading variables, and by emission permit trading variables: trades transform regional modules into a single multi-regional energy model. In TIAM-Grantham 15 different regions are implemented:

Africa (AFR)	Eastern Europe (EEU)	Middle East (MEA)
Australia and New Zealand (AUS)	Former Soviet Union (FSU)	Other Developing Asia (ODA)
Canada (CAN)	India (IND)	South Korea (SKO)
Central and South America(CSA)	Japan (JPN)	USA (USA)
China (CHI)	Mexico (MEX)	Western Europe (WEU)

Partial Equilibrium on energy markets: TIMES economic equilibrium includes three fundamental properties: linearity, maximization of surplus, and competitiveness of energy markets with a perfect foresight. These properties in turn result in two additional features: marginal cost pricing (at equilibrium, total surplus is maximized), and the profit maximization property. The perfect foresight assumption may be relaxed by assuming that some parameters are uncertain. This assumption is at the basis of the Stochastic Programming option of TIMES, that has not been used for my thesis work.

3.2.1   Time Horizon

The time horizon is divided in *time periods t*, each containing an arbitrary number of years: for all quantities such as capacities, commodity flows and operating levels, any model input or output related to the period t applies to each of the years in that period, except for investment variables, which are usually made only once in a period. The initial period is a past period where all quantities of interest are fixed to historical values: in Grantham-TIAM calibration is done up to the year 2020, while 2030 is the first period in which model optimization actually starts.

In addition to that, there is the possibility to define *time slices* within a year, so to characterize different seasons or the day/night turnover.

3.2.2 Economy

As stated before, TIMES is a *partial equilibrium model*: it simultaneously configures the production and consumption of commodities (i.e. fuels, materials, and energy services) and their prices. The price of producing a commodity affects the demand for that commodity, while at the same time the demand affects the commodity's price.

The objective function to be maximized is the *discounted sum of annual costs minus revenues*. System total cost includes both construction (investment costs), operation (i.e. fix and variable O&M, costs for domestic resource production and exogenous imports, taxes/subsidies on commodity/process activities or investments and revenues from exogenous exports) and Decommissioning. Investment costs are transformed into annual payments, taking into account capital depreciation (around 5 %) and return on investment (around 7% of capital).
In TIMES, economic equilibrium conditions determine what technologies are competitive, marginal or uncompetitive in each market, therefore it is a decision made mainly on a cost-competitiveness basis.

At the end of the linear optimization, TIMES model provides as outputs the least cost solution to satisfy energy service demands and constraints, the amount of technology investments (with capacities and related costs), the annual activities for each technology, in term of input and output, emission trajectories as well as marginal prices of energy commodities and the total discounted system cost.

3.2.3 The Reference Energy System

Within TIMES, the Reference Energy System (RES) represents all internal connections in the energy sector, being the core of this technology-rich model.
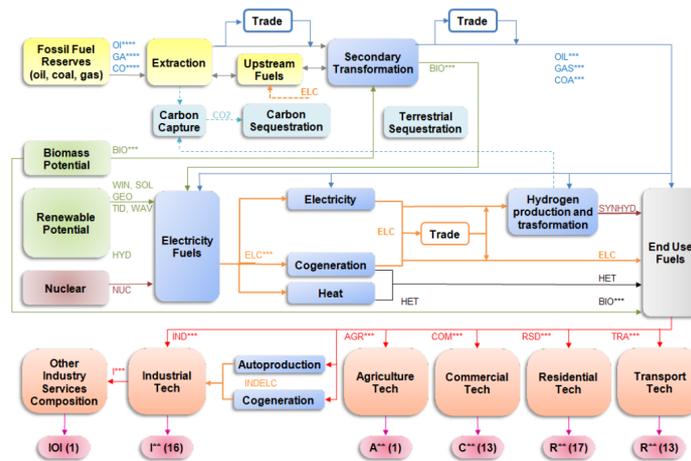

Figure 3.1: General scheme of the Reference Energy System in TIMES

The energy economic representation is based on three entities:

1. *Technologies* (also called processes), representing physical devices that transform commodities into other commodities. Processes may correspond to primary sources of commodities (e.g. mining processes, import processes), transformation activities (e.g. conversion plants producing electricity, energy-processing plants such as refineries) and end-use demand devices (e.g. cars and heating systems).

2. *Commodities*, consisting of energy carriers, energy services, materials, monetary flows, and emissions. A commodity is generally produced by some process(es) and/or consumed by other process(es). Commodities belong to five major groups: energy carriers, materials, energy services, emissions and monetary flows.

3. *Commodity Flows* are the links between processes and commodities. A flow is of the same nature as the related commodity but is attached to a particular process, representing one input or output of that technology. For instance, heating oil is a commodity, whereas heating oil for residential oil furnace is a commodity flow.

As a result, RES appears as a network diagram representing these 3 entities, as it can be seen in the following figure 3.2:
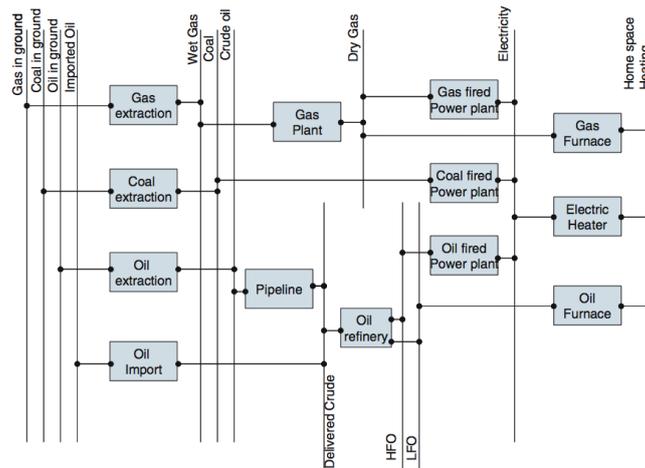

Figure 3.2: Partial view of a simple Reference Energy System

Within the RES, Greenhouse Gas emissions include both CO_2, related to energy consumption, and CH_4, from energy consumption as well as from some non-energy sectors, such as landfills, manure, wastewater or biomass burning. In addition, N_2O from energy consumption as well as from acid industries can be modeled.

All GHGs emissions are then merged into a single CO_2-equivalent emission, based on their global warming potential (GWP), and used as input into the climate module, when available. In my research work, the climate model was not adopted, and it has been preferred to set the mitigation target in terms of cumulative carbon budget (see section 4.6) rather than relying on the correlations between carbon dioxide and temperature levels implemented in this module.

TIAM is able to simulate different types of emission abatement measure, such as energy substitution within the available portfolio, improved efficiency of installed device, sequestration (CO_2 capture and underground storage, biological carbon sequestration), regulations and taxes, as well as a cap-and-trade system. Indeed, endogenous trade of all emissions is available, so to model permit trading.

3.3 Expert Elicitation

From previous modeling exercise [7, 8], it emerged that DAC deployment within IAMs is highly influenced by the choice of some key parameters, in particular the constraints on diffusion rates. These are determined mainly by modeler's choices and assumptions, therefore they are affected by a high degree of arbitrariness. In order to overcome this limitation, in the scientific community expert elicitations are often adopted to derive information about future cost reductions and deployment rates of energy technologies [64], using the results as inputs for Integrated Assessment Models.

Expert judgements are expression of an informed opinion based on their knowledge and experience about technical problems. These can complement other available data based on models' predictions, thus providing an additional source of information to inform policy-makers. It should be taken into account that experts can be subject to the same cognitive and motivational biases as all human beings. The questionnaire for the Expert Elicitation (EE) was designed according to the protocol developed within FEEM (Fondazione ENI Enrico Mattei) by Bosetti and Catenacci. This protocol was already apply to gather opinion about future prospect for solar technologies [65] and batteries [66], as well as to investigate the potential for biomass in the energy sector [67].

According to the perspective of my work, the main parameters to be elicited are the *energy consumption* (electricity and heat) for different DAC technology options, the *learning rate* that can be applied for future cost reduction, considering both Learning-by-Doing and Learning-by-Research mechanisms, the *maximum annual addition capacity*, being the amount of new DAC capacity that can be installed globally each year, and the *maximum cumulative capacity* that can be reached [$Gton_{CO_2}$/year].
In the first part of the questionnaire, experts are asked to evaluate the current technology status and the barriers to its commercial success. After that, their opinion about energy consumption of different DAC options and the expected evolution of costs in different mitigation scenarios is inquired. Finally, in the last section, some questions on diffusion pathways and externalities are discussed. The full questionnaire can be found in Appendix A.
The experts involved in the elicitation exercise are:

- Marco Mazzotti, full professor of Process Engineering at ETH, Zurich

- Nial McDowell, leader of the Clean Fossil and Bioenergy Research Group at Imperial College London

- Matteo Gazzani, assistant professor at the Copernicus Institute of Sustainable Development, Utrecht University

- Tim Kruger, programme manager of the Oxford Geoengineering Programme

The main obstacle that has been encountered during these interviews was in the different perspective when dealing to the DAC technologies: all the experts involved are technical researchers, focused on the optimization of technical details for these processes, while my perspective was the one of an energy modeler, more interested in high level of details to be able to capture the interactions of DAC plants within the overall energy system. Moreover, it was difficult to combine their research expertise with questions regarding the long-term deployment of a technology that is still at its early stage, making it difficult for them to make any predictions on how costs and technical parameters could develop.

Marco Mazzotti:
Solid Adsorbent and Waste Heat potential

Mazzotti research activity at ETH Zurich deals with adsorption-based separations and chromatography, as well as crystallization and precipitation processes. He has been coordinating lead author of the IPCC Special Report on Carbon Dioxide Capture and Storage [68] and he worked on the optimization of APS design for Direct Air Capture [29]. In the following years, he has supported researches done by the founders of Climeworks on amine-modified adsorbent, therefore his knowledge is largely focused on carbon capture with solid sorbents.

He thinks that modularity will be the effective approach to quickly scale up, as Climeworks is doing, thereby reaching significant learning effects: higher cost reduction rates can be expected for modular designs, which can be standardized and built in large amounts, than for large processing plants, as the ones based on strong base solutions. According to Mazzotti, traditional CCS technologies usually have a learning rate of 12%, so modular designs could allow even higher rates. DAC could benefit from the development of CCS, especially from the transport and storage infrastructure.
Considering the technology proposed by Lackner, he does not think that it will be deployed on a commercial scale, as few scientific evidence have been provided to support its working principle and no demonstration facility is known to have been built yet.

Nial McDowell:
Large scale, Waste Heat and Infrastructure

Nial McDowell leads the Clean Fossil and Bioenergy Research Group at Imperial College and is a member of the Centre for Process Systems Engineering and the Centre for Environmental Policy. His research interests are highly interdisciplinary, focusing on integrated multi-scale modelling of low carbon energy systems and their dynamic interactions across varying length and time scales.

McDowell thinks that the APS design, then further developed by CE accordingly, could lead to important economies of scale, as it results in big-scale power plants with a capture capacity around 1 Mt/yr. Differently, Climeworks design is focused on a smaller scale, being a modular setup: this may have an impact on transport costs which are expected to be higher than for concentrated, large scale plants. Moreover, the possibility to use waste heat from industrial processes to operate amine-based plants will reduce their location flexibility as they need to be sited close to the heat source. He agrees that artificial tree won't be really an option for DAC, resulting more a "dream" technology: there is still a lack of data and scientific calculations published, while only low purity CO_2 can be obtained, that cannot be used directly for storage.

Matteo Gazzani
Tecnical Considerations on DAC Options

Matteo Gazzani's research focuses on energy systems, integrating technical aspects from chemical, mechanical and electrical engineering to enable the deployment of new technologies for clean energy production. He is particularly interested in CO_2 capture and storage, enhanced gas separation, optimization of decentralized energy systems, water-energy nexus, and decarbonization of energy intensive industries. Recently, he worked on the optimization of DAC systems, both liquid scrubbing based on NaOH solutions and amine-based sorbents. He is the one that tried to fill out the questionnaire in the most extensive way.

According to Gazzani, currenlty there are only two chemical approaches for Direct Air Capture, namely adsorption using solid materials that selectively bind CO_2, or absorption using liquid solutions containing chemical sorbents with strong CO_2 affinity ($CaOH_2$, NaOH, KOH, $NaBO_2$, always as a combined double-loop).
Considering aqueous solutions, he thinks that advances are still needed to make it more competitive, through engineering and applied R&D but the potential for further efficiency improvement is quite limited as well known and mature technologies are employed. The main barriers for this option are the complexity of the double-loop process, the high number of steps and equipment needed and the high energy needs for regeneration. According to his perspective, APS estimates for costs can be still considered as a reliable benchmark.
When moving to amine-based processes, the specific barriers identified are the choice of materials and their costs, as well as sorbent degradation, that can have high impact in overall performances. The advantage of this approach is that regeneration temperature cannot go above 120°C otherwise leading to amine evaporation, containing energy consumption. Today there is sufficient knowledge to build demonstration plants, as both GT and Climeworks have their own pilot, but in parallel basic R&D needs to continue to fully assess the potential of improvements for this technology. The figures provided by GT and Climeworks for energy consumption are reliable and consistent with his optimization results. Moreover, both Climeworks and GT cost estimates for the near-term (200 and 150 $/ton, respectively) may represent a realistic price target.

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When discussing about the potential diffusion for DAC on the market, Gazzani's opinion that it is not a technological problems, as the technology itself is already available, but the key aspect will be in the price that is given to carbon dioxide (i.e. the carbon price). Therefore, the main barrier is from a governmental and policy perspective: someone has to support the deployment of this technology and a strong political commitment is needed, being responsible (and paying) for DAC installation.

Tim Kruger:
Focus on co-Benefits and co-Products

Tim Kruger leads a group across Oxford university exploring proposed geoengineering techniques and the governance mechanisms required to ensure that any research in this field is undertaken in a responsible way. Moreover, he is developing a new interesting approach to capture CO_2 from the air, coupling a lime kiln for Ca carbonation with a Solid Oxide Fuel Cell [69].

Kruger underlined that many studies concluded that mitigation options as CCS can only become economically viable when a commercial value is given to the output products, in this case the stream of captured CO_2 emissions, thus overcoming challenges related to high cost and immature technologies. Therefore, he thinks that the focus should move to CCU (Carbon Capture and Utilization) processes, using CO_2 for instance to produce transport fuels such as methanol. These application would require a substantial amount of energy, so that it results more profitable and convenient to use curtailed energy from renewable sources to directly operate DAC power plants, and then maybe transform CO_2 into valuable products: the advantage is that DAC plants can operate in flexible manner, adapting to the availability of surplus electricity from renewables. He appears to be quite skeptical about the future potential of all designs proposed for DAC so far, as they include only costs and no stream of revenues from valuable output products.

Even if the final outcome was different from the expected one, as very few experts were able to fill in the questionnaire and to give their opinion about the key elicited parameters, the Expert Elicitation exercise results very helpful in modeling DAC technologies within TIAM.
Generally, experts agreed that using strong base solutions will lead to large scale plants, while amine-based sorbent will have a modular design paradigm with higher learning rates as they can be standardized and built in large amounts. This can have an impact on transport and storage cost. Moreover, all of them assent that artificial tree does not represent a realistic DAC option as for now, due to the lack of data and scientific calculations, but it should be regarded more as an *extreme and futuristic* alternative.

Chapter 4

DAC Implementation in TIAM

In this chapter, the implementation of different options available to realize Direct Air Capture in TIAM will be discussed, considering techno-economic parameters (Section 4.1), growth constraints (Section 4.2), sequestration (Section 4.3), the competition with other Negative Emission Technologies (Section 4.4) and their environmental footprint (Section 5.5. Information found in the literature about currently available technology are combined with the results from the Expert Elicitation and historical comparisons to determine expected diffusion rate and cost reduction pathways.

4.1   Direct Air Capture Technologies

As it has been described previously, in TIMES each process can be modeled by defining a number of input and output commodities coming in/out of a *box*: these flows can be then related defining input/output coefficients or efficiencies. Input flows represent the energy required for the capture process, both in term of electricity and heat: heat can be supplied in a number of ways according to the different technology analyzed. The main output flow is the stream of captured CO_2 to be sent to storage sites: according to TIAM framework, a new commodity was created, so to differentiate the carbon dioxide captured by DAC technologies with respect to other sequestration options such as CCS in power plant and industrial sector or other NETs. In this way, DAC can be modeled and analyzed more extensively and independently from other mitigation strategies, focusing for instance on the location flexibility of these plants that could reduce transportation costs (see Section 4.3).

According to the state of the art presented in Chapter 2, I decided to differentiate across three technology options: the one based on hydroxide solution, on amine sorbents and the artificial tree[1], referred to as DAC1, DAC2 and DAC3 respectively.

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[1]Other solid adsorbents have been discarded as still too far from actual implementation.

1. *Strong Base Sorbents - DAC1*

   NaOH, KOH, $Ca(OH)_2$, based on APS design and CE demonstration plant.
   This technology has been explored more extensively than other technologies and can borrow some components from existing processes (e.g. paper industry), so that it would be available earlier (already in 2020), but the potential for cost reduction will be limited, with a higher floor cost than for other categories.
   It requires a large amount of energy for regeneration (thermal need $\sim$ 8 GJ/ton), that will be provided with natural gas so to reach temperature above 800°C.
   Major expenses are related to capital costs, with the packing material being the biggest contributor, according to APS breakdown [20].

2. *Amine-modified Solid Adsorbent - DAC2/DAC21*

   based on Goeppert research [25], and pilot plants by GT and Climeworks.
   As the temperature during regeneration is about 80-100°C, it will have lower energy consumptions and the heat could be provided using waste heat from industrial processes and power plants: this option is applied to the technology DAC2, while DAC21 processes use low-temperature heat made on purpose.
   These plants have a modular setup, that allow to reach a mass production paradigm, thus enabling further cost reduction in the future. As the scrubber required for this category of sorbent is simpler and cheaper than for hydroxide solutions, capital costs only account for a smaller fraction of the total, while operating costs will be much higher, given also stability and degradation issues related to amine-based solvents. In order to model sorbent degradation, the lifetime of these plants has been reduced to 15 years, while for others it is 20 years.

3. *Artificial Tree - DAC3*

   based on the design proposed by Lackner [31].
   Only electricity needs to be supplied to these plants for sorbent regeneration. As low purity CO_2 is obtained, an additional separation step similar to traditional CCS needs to be added at the end, rising energy and cost estimates.
   All the experts agree that this technology won't be delivered in the next future, given the high degree of uncertainty about it (see Section 3.3), therefore I decided not to include this process in the main runs and to leave it as an extreme option to be investigated, due to the favorable energy needs and costs claimed.
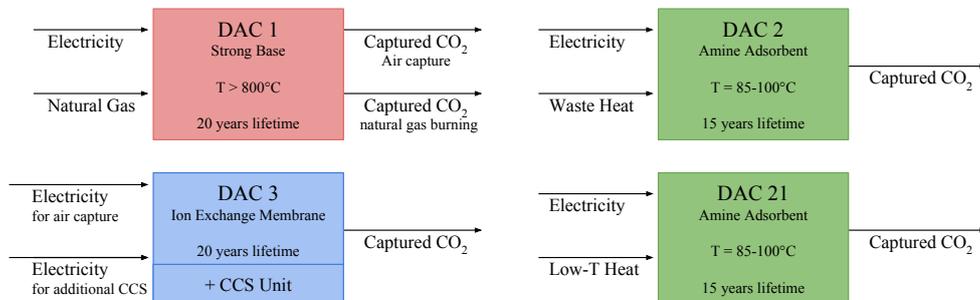

Figure 4.1: DAC processes representation within TIAM, with input/output commodity flows

4.1.1 DAC Energy Requirements

For each technology category, input commodity flows have been defined to represent the energy required for the capture process, considering the consumption specific to each ton of CO_2 captured. According to the literature values, both a *high* and a *low energy scenario* have been examined for the robustness analysis, so to represent the uncertainty related to this parameter: with a conservative approach *high energy needs* are considered as the base case, while more optimistic estimates are applied in the sensitivity. The final values considered are summarized in table 4.1.

Code	Technology			Electricity [GJ/ton]	Heat [GJ/ton]
DAC1	Strong Base $Ca(OH)_2$/NaOH/KOH	high	(APS, 2011)	1.8	8.1
		low	(Keith, 2018)	1.32	5.25
DAC2	Amine-functionalized adsorbent (waste heat)	high	(Climeworks)	1.1	7.2
		low	(GT)	0.7	4.3
DAC21	Amine-functionalized adsorbent	high	(Climeworks)	1.1	7.2
		low	(GT)	0.7	4.3
DAC3	Ion Exchange Membrane	extreme scenario		1.14 + 1.33 for CCS	-

Table 4.1: Energy Requirements for different DAC technologies

In order to differentiate energy requirements across technologies, heat is provided to DAC1 by burning natural gas , while for amine-based adsorbents it may come from the recovery of waste heat (DAC2), or using intentionally produced heat (DAC21), supplied mainly through biomass-based processes. The implementation of a new commodity representing waste heat recovery will be discussed further in details in next section.

It should be noted that the design proposed by the APS report requires an additional CCS unit to capture the CO_2 emitted by burning natural gas to supply the high-temperature heat needed for the regeneration process. In order to account for combustion emissions in TIAM, fuel-based emission coefficients are defined, so to link emissions directly with the amount of fuel burnt throughout the energy sector. This means that for each PJ of natural gas burnt, the model accounts directly for 56.1 kton of CO_2 emitted into the atmosphere. Therefore, the amount of CO_2 captured from the kiln needs to be defined as an additional output flow equal to 53.2 kt/PJ, that corresponds to a capture efficiency η_{ccs} close to 95%, according to how reference NGCC plants with oxyfuel CCS are already implemented in the model.

Moreover, when defining Artificial Tree systems, the electricity input needs to consider both for the energy needed by the process itself (1.14 GJ/ton, according to [31]), and by the additional CCS unit required to produce a stream of high purity CO_2 that can be send to sequestration sites. As discussed previously, the output of this moisture-swing driven system is a stream with a CO_2 fraction close to 5%, which is

similar to the concentration in flue gases from natural gas based power plants. In order to compress it and send it to storage site, the additional capture requires 1.33 GJ/ton [70], impacting also on the overall cost (see Section 4.1.3).

4.1.2 The Use of Waste Heat

As it has been highlighted previously, a big advantage for plants employing amine-modified adsorbents may come from coupling DAC processes with industrial waste heat so to reduce the impact in term of energy (heat) requirements and related costs. Note that this is already being implemented both in Climeworks' and Global Thermostat's plant, therefore it is key to understand where this heat may be recovered from and which is the availability at a global scale.

In order to include this aspect within TIAM framework, a new commodity representing waste heat was added to the model: it can be recovered both from energy-intensive industrial processes (e.g. pulp and paper, iron and steel, chemicals, glass, cement) and from the power sector. As the waste heat potential will be limited, two distinct processes to represent amine-based DAC have been defined, namely DAC2 and DAC21: these will have different input commodities to represent heat supply (see Figure 4.1), but same cost and technical parameters: in this way, the model is free to install as much capacity of amine-based DAC plants through DAC21 technology, with no constraints deriving from the available stream of the waste heat commodity.

1. Waste Heat from Industrial Processes

The recovery and re-use potential of industrial waste heat is determined by multiple factors, including the characteristics of waste heat sources and sinks, their compatibility in terms of temperatures, capacity, timing or location, the costs and efficiency of available recovery technologies and energy/carbon prices that could make it attractive. To understand these factors, databases of industrial waste heat sources, sinks and heat recovery technologies needs to be built based on literature data and discussions with industry partners.

For my research work, I have been referring to the report developed by Ecofys and Imperial College London [?]: it examines a number of heat intensive industrial sectors, such as refineries, iron and steel, ceramics, glass, chemicals, food and drink and pulp and paper industry. A database was created describing archetypal characteristics of waste heat sources and heat sinks at 73 largest UK industrial sites: the report describe the overall waste heat potential available, as well as the technical and economic potential that can be actually extracted from these sources based on existing technologies. Referring to these data, I have identified the sectors where the new commodity may be pulled out and for each of these a benchmark for the recovery process has been established, defining the production of waste heat as a fraction of the energy input, with this recovery factor changing according to the industry[2], as it can be found in Table 4.2:

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[2]Note that for Other Industries category, the recovery factor has been defined as the average of other sectors' values.

Sector	Recovery Factor	
	Min	Max
Iron and Steel	35%	32%
Pulp and Paper	30%	
Chemicals	20%	
Cement and Glass	40%	30%
Non-ferrous	35%	32%
Other Industries	30%	

Table 4.2: Waste heat recovery factor for different industrial sectors.

I made the simplifying assumptions that that fraction of rejected heat can always be retrieved, while in reality, recovering is furtherconstrained by the ability to re-use it and its economic value [71]. Moreover, thermodynamic limits should be considered, being represented by the match between source and sink temperatures. As for our case the sink is represented by amine regeneration processes, taking place at temperatures below 120°C, only industrial heat flows with temperature equal or higher than 140°C examined in the report have been taken into account.

Considering the processes already present in TIAM in the industrial sector, the fuel used as input and the installed capacity in baseline scenarios, the waste heat commodity was added to the following categories of industrial processes:

1. Gas-fired processes for production of process heat and steam cross different energy-intensive industries: iron and steel, cement and glass, chemicals, pulp and paper, as well as non-ferrous metal production and food (other industry). Other fuels beside natural gas have not been included, given that their role won't be relevant in the second half of the century (if not coupled with a carbon capture unit) in stringent mitigation scenarios, due to a transition to low carbon sources.

2. CCS processes, burning both natural gas and coal in all sectors listed before.

3. For the sector including other industries (OI), processes burning different fuels than natural gas have been considered (e.g. biomass, coal and oil), given that they have still a significant capacity installed up to the end of the century.

4. Additional processes in the iron and steel sector not devoted to the production of process heat or steam (e.g. electric arc furnace), burning natural gas.

It should be noted that a cap has been put on these industrial processes connected with waste heat, to avoid the model to overinstall them only to provide the heat commodity needed by DAC plants: a constraint was implemented based on the capacity installed in a baseline scenario, with a mitigation target consistent with 2°C.

When exploiting waste heat for running DAC2 plants, the flexibility in location is reduced as the plant should be built close to the heat source, not to the storage site. In order to account for this aspect, the technology DAC2, that is the one using waste heat commodity as input will have the same output commodity of normal sequestration technologies, and the same transport cost.

2. Waste Heat from the Power Sector

It has been decided to add a flow of heat also from some renewable power plants, such as nuclear and solar thermal. It should be noted that in the first case it is actually waste heat that can be recovered from the steam cycle in addition to the main electric output, while in the latter case, heat is produced as in Combined Heat and Power (CHP) units, thus slightly reducing the main electric output.
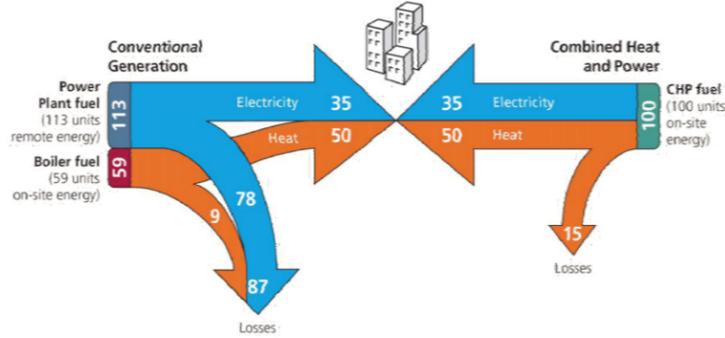

Figure 4.2: Combined Heat and Power design, from [72].

Concentrated Solar Power

As explained before, heat can be recovered from Concentrated Solar Power (CSP) used as CHP units, therefore a new process has been implemented to differentiate it from traditional CSP plants only producing electricity. This technology will use a steam power plant with back pressure configuration, that means the electricity output is reduced increasing the temperature at the outlet of the steam turbine, so to have sufficient high temperature to allow heat recovery at the condenser, obtaining a useful stream of heat[3]. Therefore, CHP solar plants will have lower electrical efficiencies with respect to the ones producing only electricity, going from η_{el}=20% to 15% [73]. Considering a fixed energy input to the plant, the output flow of electricity will be reduced from the initial unitary value to 0.7 for each capacity unit of this process, according to:

$$ELCC_{out} = \eta_{el,CHP} \cdot Q_{in} = \eta_{el,CHP} \cdot \frac{1}{\eta_{el,only}} = \frac{15\%}{20\%} = 0.7$$

In order to compute the stream of waste heat commodity that can be obtained for each unit of electricity produced, a *Recovery Factor* is defined:

$$RF = \frac{Q_{wst}}{W_{el}} = \left(\frac{1}{\eta_{el}} - 1\right) \cdot \eta_t = 4.53$$

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[3]Remember that for DAC2 application, heat is needed at around 100°C, therefore the source should be at a higher temperature to allow a sufficient ΔT of pinch point in the heat exchanger.

where η_{el} is the electric efficiency of the Rankine cycle considered (i.e. 15% for a CHP-CSP plant) and η_t is the waste heat recovered from the cycle, equal to 80% of the heat output at the condenser [73]. The recovery factor is then multiplied for the electricity output (0.7), to rescale it considering a capacity unit of this technology. Costs are the same, but they will be allocated to a reduced electricity output, so that this technology will result more expensive for the model with respect to traditional CSP plants. As both electricity and heat are primary output commodity (not auxiliary ones as waste heat coming from industrial recovery or nuclear plants), the overall capital cost is equally split among these two output flows, therefore waste heat may result slightly more expensive for the model if coming from solar plants, but it would have the additional benefit of producing electricity at the same time.
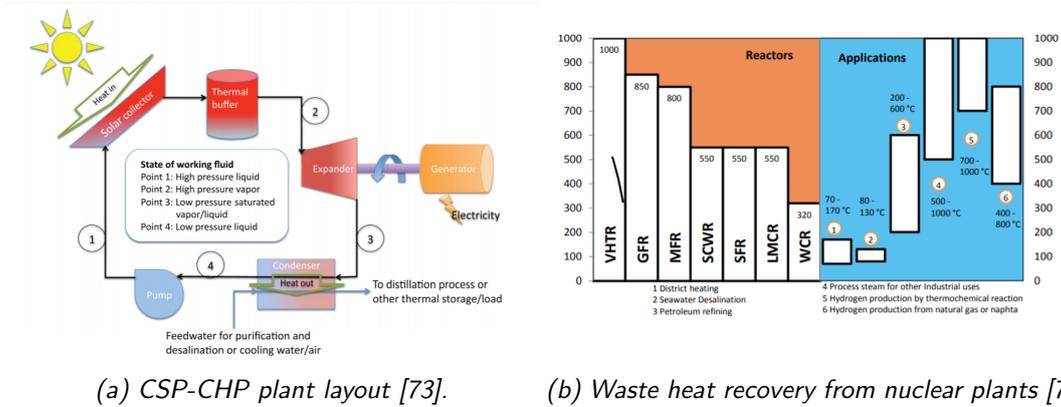

(a) CSP-CHP plant layout [73].   (b) Waste heat recovery from nuclear plants [72].

Figure 4.3: Waste heat from power sector, solar thermal (a) and nuclear plants (b).

Nuclear Power

Current efficiency of electricity generation from nuclear plants is about 33%, which is rather low since about two-thirds of the energy in the fuel is lost and dissipated to the environment as heat. Using waste heat recovery technology to capture a significant proportion of this lost heat, the efficiency could be increased, with cogenerative systems achieving efficiencies of 60 to 80% producing at the same time electricity and thermal energy. Other applications for this recovered heat may be seawater desalination, hydrogen production, district heating/cooling, or energy-intensive industrial processes [72]. All existing nuclear reactor types can be used in cogeneration mode without reducing the main electrical output, as the temperature of the dissipated heat is sufficiently high to produce the heat needed by DAC plants.

Therefore, n TIAM, all existing nuclear processes have been redefined, adding waste heat as an auxiliary output commodity, The overall efficiency of cogeneration plants is usually about 80%, including an electrical efficiency η_{el}=30-35% and a thermal efficiency around 50% [72]. Therefore, for each PJ unit of electricity in output, 1.3 PJ of waste heat may be obtained, according to:

$$RF = \frac{Q_{wst}}{W_{el}} = \frac{\eta_{el}}{\eta_{th}} = 1.3$$

4.1.3 DAC Cost Assumptions

Considering the cost estimates discussed previously in Section 2.8, it has been decided to include both a *high* and a *low cost scenario* according to the different values found in the literature (see Table 4.3). Generally, scientific papers are used as a reference to determine the higher bound, while estimates by companies are used to determine the low cost scenario and/or the floor cost that can be reached, being more optimistic on the expenses associated with these capture processes. In particular, *floor cost* reflects the long-term target that could be achieved in the future, and are used to set a lower bound for cost reduction when learning effects are taken into account (see Section 4.1.4). It is important to underline that operating expenditure reported in the table do not include energy costs, as these are determined endogenously by the model that assigns a price to each commodity flow entering a process. All the costs shown have been then adjusted to $\$_{2000}$ considering the inflation factor of the corresponding year. Note that capital cost (i.e. CAPEX) sometimes is indicated as an annualized value in [\$/ton], while other reference may include it as an investment cost in M\$ for the reference size of the power plant considered. In order to move from the investment cost to the annual payment spread over the plant lifetime, the *Capital Recovery Factor* (CRF) needs to be computed, according to the formula:

$$CRF = \frac{\left(1 - r_s(t)\right)}{\left(1 - r_s(t)^{T_{life}}\right)}$$

where $r_s(t)$ is the technology-specific discount factor, and T_{life} is the technical lifetime of the plant[4]. The discount factor can be computed from the technology-specific discount rate $d_s(t)$, being equal to the 10%, as we are dealing with immature technologies:

$$r_s(t) = \frac{1}{1 + d_s(t)}$$

Code	Technology			CAPEX		OPEX
				[M\$]	[\$/ton]	[\$/ton]
DAC1	Strong Base	high	(Mazzotti, 2013)	(2060)	220	76
	Ca(OH)$_2$/NaOH/KOH	low	(Keith, 2018)	1146	(140)	42
		floor cost	(Keith, 2018)	700	(75)	27
DAC2	Amine-functionalized	high	(APS, 2011)	(750)	90	260
DAC21	adsorbent	low	(Climeworks)	(430)	50	150
		floor cost	(GT)	(110)	13	37
DAC3	Ion Exchange	base	(Lackner, 2009)	(800)	85	100+20
	Membrane	floor cost	(Lackner, 2009)	(80)	8.5	21.5+10

Table 4.3: Cost assumptions for different DAC technologies

For DAC1, results from Mazzotti optimization on the APS design [29] have been considered for the high cost scenario, with an overall cost around 300 \$/ton (without energy), while the latest paper by Keith and Holmes issued in June 2018 provided

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[4]As DAC2 plants have a shorter lifetime, the CRF results higher than for DAC1 and DAC3 plants (0.120 and 0.107 respectively), meaning that capital expenses are allocated over a shorter span of time.

the values both for the low cost scenario (overall price around 180 \$/ton) and the target for further cost reduction, around 105 \$/ton. Both these references provide a detailed breakdown for cost assessment, therefore it is possible to differentiate capital and operational expenditure, as well as individual cost components. Note that this technology is characterized by a higher floor cost to reflect its limited potential for cost reduction, as it is employing processes and equipments already well-known and developed in other sectors.

Capture plants based on amine-modified adsorbents (both DAC2 and DAC21) are characterized by higher operational costs due to frequent sorbent replacement because of degradation. As it is difficult to find exhaustive cost assessment for this technology, it has been decided to start from APS estimate for the overall capture cost (350 \$/ton without energy), but switching the fraction allocated to OPEX and CAPEX (74% and 26% respectively), so to reflect the peculiarity of this technology option. The low cost scenario is defined according to Climeworks estimate (200 \$/ton, keeping the same allocation for capital and operational expenditure used before), while the floor cost of 50 \$/ton is the one claimed by Global Thermostat.

Considering ion exchange membrane (DAC3), all information about costs are derived from the only scientific reference available by Lackner [31]: capture cost is estimated to be about 200 \$/ton, out of which 15 \$/ton are related to electricity consumption, with an expected reduction of capital expenditure of 90% (from 200 000 to 20 000 \$ per unit). The cost target indicated is equal to 30\$/ton, being by far lower than other estimates and therefore strongly criticized as too optimistic. It should be noted that a supplementary operational cost is considered, to account for the additional concentration process required to obtain sufficient purity CO_2: being this step similar to CCS from natural gas plants, it will require 20 \$/ton today [70], with a 50% reduction expected in the future thanks to its technological development.

4.1.4 Cost Reduction and Learning Effects

The characteristics of future technologies are inevitably changing over the next decades due to technological learning: generally learning curves are applied both to energy requirements and costs, resulting a function of time, cumulative capacity, and R&D investment. Significant cost reduction may result from R&D before a technology enters the market, as well as further rebates can take place after market introduction, through learning-by-doing, economies of scale, continued research and maturing supply chains. Generally, this applies also to energy requirements through efficiency improvements, but I decided to keep them fixed in time, investigating model sensitivity to high and low values.

Given that it is not possible to foresee exactly the evolution of costs, three different approaches may be used when working with IAMs:

1. Assume *no technological change* to examine whether, with stock turnovers, cur-

rent technology characteristics are sufficient to meet energy system goals.

2. Use *Exogenous Technical Learning*, that is an exogenous forecasts of how technologies' costs may develop in the future. Cost reduction depends only on the time elapsed and may thus be specified outside the model. It is possible to forecast such changes as a function of time according to historical comparison with similar technologies, and thus to define a time-series of values for both capital and operational expenditure:

$$C(t) = C(t_0) \cdot (1-a)^{(t-t_0)}$$

where a represents the annual cost reduction rate. This value may differ according to the technology examined, and benchmark values can be identified considering past cost reduction pathways achieved in the energy sector and beside it.

3. Use *Endogenous Technical Learning (ETL)*, meaning that the future cost parameters are no longer a function of time alone, but depend on the experience acquired and the knowledge stock accumulated around that technology by installing more plants. Therefore, future costs typically depend on the cumulative capacity installed, or equivalently on cumulative investment decisions taken by the model, which are unknown before running the model. For DAC, the capacity installed may be represented by the amount of CO_2 captured, accounting also for other sequestration options, such as CCS and BECCS, given the similarities among these technologies and the knowledge spillovers that could be achieved. This mechanism is also named *Learning-by-doing* (LBD).

$$C(t) = C(t_0) \cdot \left(\frac{Cap_t}{Cap_{t_0}} \right)^{-b} \qquad pr = 2^{-b}$$

where b is the learning index, representing the speed of learning: usually it is defined starting from the progress ratio pr, that defines the rate at which cost decline when the cumulative capacity/knowledge stock is doubled.

In TIMES, it is possible to represent endogenous learning for the unit investment cost of technologies, but this requires a Mixed Integer Programming formulation, instead of standard linear optimization. Moreover, the model does not allow to include knowledge spillovers, linking cost reduction for one technology to the capacity of other similar technologies. Hence, due to TIAM mathematical structure, the impact of ETL formulation results not suited to the purpose of my modeling exercise.

Therefore, I decided to apply a simple exogenous cost reduction, investigating different reduction rate a, with the possibility to assess the impact of a *pseudo Learning-by Doing*, built exogenously according to the amount of CCS and DAC installed in previous model runs, in an iterative way. As the impact of different reduction rates appeared limited (see section 6.1), I decided not to go further in this direction with additional complexity, given the restricted contribution to the overall study.

In order to investigate the impact of learning mechanisms for the deployment of DAC technologies, exogenous cost reduction rate (defined as annual percentage) have been derived from historical recorded data, using the dataset developed by Farmer [74] that covers a wide range of technologies, not only in the energy sector but in also hardware and chemical industries. Historical values are summarized in table 4.4.

Sector	Min	Max	Avg
Energy	4%	10%	6%
Chemical	1%	11%	6%
Hardware	30%	44%	37%
Consumers Good	2%	8%	5%

Table 4.4: Historical annual Cost Reduction Rate in different sectors, according to [74].

As a baseline case, it has been used a 6% annual reduction, which is the average historical value both for the energy and the chemical sectors, which are the ones more related to DAC technologies. Then, two extreme annual rates have been considered for sensitivity analysis, equal to 15% and 1%.

Cost Reduction	Min	Base	Max
DAC	1%	6%	15%

4.2 Diffusion and Expansion Constraint

As TIAM-Grantham operates on a least-cost basis, it is likely that the cheapest technology in any sector will be deployed without any feasible limit, with a pattern that cannot be considered realistic in the near-term. Therefore, technology growth constraints are frequently employed in such models, in order not to have technology penetration pathways unrealistically rapid. Expansion constraints can be modeled as:

- *Maximum Capacity Installed*, that is a ceiling on the extent to which a technology can be deployed globally. This corresponds to an absolute constraint:

$$DAC(t) \leq DAC_{max} \quad \forall t$$

- *Annual Growth Rate* on cumulative capacity installed, corresponding to a relative constraint on the marginal increase:

$$DAC(t+1) \leq DAC(t) \cdot \alpha$$

where α is the coefficient that limit the growth of a technology per each time period (i.e. the annual growth rate, as it has been investigated by Iyer [75]).

- *Logistic Growth* for the cumulative capacity, to model the diffusion pathways of any technology in the market, as it has been investigated by Wilson [76]

$$f(t) = \frac{L}{1 + e^{-b(t-t_0)}}$$

where L is the maximum installed capacity

It has been decide to focus on the first and second approach: logistic curves are generally used to fit historical diffusion pathways [76] and to analyze ex-post the lifecycle of energy technologies in term of up-scaling, formative phase and saturation, while annual growth rates appear more appropriate to constraint the growth up front, so to avoid unrealistic scenarios.

4.2.1 Global Maximum Capacity Installed

One of the most important parameter determining the deployment of DAC technologies in TIAM is the maximum capacity allowed, representing the physical scale reached in the future in terms of Gton of carbon dioxide captured via DAC plants[5]. Indeed, the model treats Direct Air Capture as a backstop technology, over-installing it in the last decades of the century to be able to meet the mitigation target imposed. In order to reduce the arbitrariness related to the choice of this parameter, a number of past modeling exercises and comparative assessments for NET deployment have been considered.

Within the Climatic Change special issue on negative emissions [7], Carbon Dioxide Removal is contemplate to reach a scale of 15 to 30 $Gton_{CO_2}$/yr by 2100. These estimates have been derived both using IAMs to investigate CDR deployment and applying ecology, carbon-cycle science and chemical engineering to assess the theoretical potential for these technologies. Generally, these second type of assessments estimates CDR to reach level between 10-15 $Gton_{CO_2}$ annually by the end of the century [77, 78, 21], while the limited evidence available from long-term IAM-based studies are at higher end of this range, around 40 $GtCO_2$.
Considering biophysical and economic implications related to different levels of NETs implementation consistent with a 2°C target [3], it has been determined that the maximum level of DAC deployment would correspond to 10 Gt/yr removals in 2100.
Recent assessments of costs and potential for negative emissions [12] agree that DAC deployment can reach level of 5 Gt/yr in 2050 and 10 Gt/yr by the end of the century. Potential uptake up to 40 Gt/yr may be possible if constraints such as environmental side-effects and land demand can be proven unjustified or able to overcome [79]. Considering past IAMs studies integrating DAC as a CDR option [4, 5, 6, 8], this technology results able to scale rapidly reaching a capture capacity around 35-40 Gt/yr.

Therefore, I decided to put a cap equal to 5 Gt/yr as a mid-term constraint in 2050, while the extent reached in 2100 is set to be equal to 35 Gt/yr for the base case, being consistent with the cumulative capacity usually reached with IAM-based

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[5]Note that this corresponds to the value of L in a logistic function profile

modeling studies. Given the influence of this parameter, the impact of a different cap will be examined through sensitivity analysis, setting the global extent equal to 10, 20 up to 50 Gt/yr at the end of the century. Note that the same overall capacity is applied as global potential for all CDR when multiple NETs are in competition, as it will be explained later in Section 4.4.

Maximum Global Capacity [Gt/yr] in 2100	Min	Base	Max
DAC	10-20	35	50

4.2.2 Annual Growth Rate

Several studies tried to assess the feasibility of future diffusion pathways for low-carbon technologies to meet ambitious climate targets, given limitations that may come from institutional, behavioral and social factors difficult to be quantified *a priori* [75, 80, 81]. In particular, historical comparisons have been extensively used in the literature to derive information about 4 key parameters: the *annual growth rate* [75], the *logistic growth profile* [76], *(exogenous) cost reduction* [74], and *learning rates* for ETL [82].

I decided to consider historical diffusion rate for a range of technologies, not only in the power sector, as a benchmark to constrain the deployment of DAC, so to avoid unfeasible scenario results. Indeed, DAC processes leading to a mass production paradigm (i.e. DAC2/21 ans DAC3) may have similarities with the diffusion of other technologies in the past, in the energy sector (e.g. solar PV, batteries), as well as in electronics/software world. I have specified the maximum expansion rate per year to be applied to the DAC capacity, according to the equation below. As no plants for air capture are present at the beginning of the simulated time horizon, an initial seed needs to be defined so to start the deployment, equal to 1 Mton of global capacity for the first year: this value reflects the reference scale identified by the APS report.

$$DAC(t+1) \leq DAC_t(1+r) + seed$$

where r is the imposed annual growth rate, that is being defined according to the survey of historical average growth rates made by Iyer et al. [75] over a range of technologies and products. The highest rates experienced in the past are about 11-19% for nuclear plants, 15% for flue gas desuplhurization systems and 20% for wind in Denmark.
DAC growth is therefore constrained to a 20% annual rate as a baseline assumption. As this results to be a binding constraint in determining the overall deployment of DAC within TIAM, sensitivity analysis is required to explore the impact of higher and lower values, equal to 30% and 10-15% respectively.

Annual Growth Rate	Min	Base	Max
DAC	10-15%	20%	30%

4.3 Transport and Storage

Carbon dioxide can be transported by pipelines, ships, and road tankers: transportation via pipelines results to be cost-effective for large quantities (> 1-5 Mt/y) and distances (> 100-500 km), therefore it is the most diffuse option nowadays [83].

As CO_2 is transported in a supercritical state, with a density ten times higher than the one of natural gas, carbon dioxide piping requires less energy. Moreover, operation records for CO_2 pipelines show low rates of carbon leakage and no major safety concerns, even if H_2S and SO_2 impurities can increase the risks associated to leakages. Transportation costs via pipeline can be related to the infrastructure costs (pipelines construction, pipe coating, protection system), as well as costs of allowances, surveillance and expert supervision. According to several sources, two variables mainly affect the transport cost: the mean pipeline length and the average CO_2 mass flow rate [84, 85, 86].

The largest pipeline transports several Mt of CO_2 over 800 km in the US. In order to reach a significant level of deployment for CCS and other sequestration technologies, the IEA projects that Europe, China, and the US may need a transportation capacity in the order of some Gt of CO_2 per year by 2030 [83].

After transportation, CO_2 can be stored in geological structures such as deep saline formations, depleted oil and gas reservoirs (with or without enhanced oil recovery), and deep, unmineable coal seams. As only few demonstration projects have been done so far, more experience is needed to understand the underground behaviour of the injected CO_2 also in term of leakage and to characterize the geological formations for large-scale, safe and long-term storage.

Deep Saline Formations - Aquifers
Deep saline formations offer the largest storage potential, which is estimated between 1000 and 10 000 Gt [83]. Saline aquifers consist of water-saturated sedimentary rocks (e.g. sandstone or carbonate): in open aquifers, water circulates on a geological time-scale and rocks are permeable enough for fluids to be injected. In closed aquifers water is confined by non permeable layers, usually with dissolved solids, thus they are not suitable for sequestration.

Ttrapping mechanisms include a free phase at the top of the aquifer, CO_2 being trapped in the pore space, CO_2 being dissolved in waterand precipitated mineral carbonates. Anthropogenic damage of the cap rock (e.g. wells) may cause leakages, but more research is needed to understand rock sealing, CO_2 geochemical transport and the impact of seismic activity.

Oil and Gas Field, with Enhanced Recovery
Carbon dioxide is the second most used fluid for Enhanced Oil Recovery (EOR), following steam, once primary production (driven by reservoir pressure) and secondary production (by water flooding and pumping) have been applied. Indeed, EOR can extract from 5% to 20% of the original oil in the reservoir , with an additional 0.1-0.5

ton of oil per ton of injected CO_2. In the next 15-25 years, preliminary estimates suggest that around 30 Mt CO_2 per year could be used for EOR, with a potential of 5-6 million barrels per day by 2030 [83]. This can be further incentivized by increasing oil prices and the availability of CO_2 transportation infrastructure: the cost of EOR-based carbon storage is currently estimated at 20-30\$/tCO2 and it is largely offset by the oil production revenue. A similar practice can be applied also to gas field, but turns to be less profitable and more expensive. Besides enhanced recovery, depleted oil and gas fields offer low-cost opportunities for CO_2 storage as facilities and wells are often in place, and the geological characterization of the site is already available.

Estimates of this storage potential range from a few $Gton_{CO_2}$ to several hundreds, mainly located in Middle East countries, Russia, Europe, North America, China and Venezuela. There are 400 sites worldwide where CO_2 emitting sources and depleted oil fields are within a distance of 100 km with a total storage capacity of 0.5 Gt/yr, .

Unmineable Coal Seams

Unmineable coal seams are the ones too deep or too poor for commercial exploitation. CO_2 storage can help in releasing methane that is absorbed into coal pores (i.e. Enhanced Coal Bed Methane recovery - ECBM), with coal being able to absorb two moles of CO_2 per mole of CH_4 released.

ECBM resources are mostly located in North America, China, Russia, India, South Africa, and Central Europe. The global potential is estimated to be between 100 and 200 Gt [83]. Few small demonstration projects are currently in operation or planned.

Other Storage Options

Few other storage options have been investigated: salt caverns have limited capacity and shallow depth, abandoned mines are usually unsuitable due to leakages, oil and gas shales have shallow depth and low permeability, basalt formations have low permeability and porosity, while mineral carbonation, which is based on CO_2 reaction with Mg and Ca silicates to form carbonates, involves a huge amount of materials.

Ocean storage has been proposed recently as a favorable alternative, but lot of concerns have been arisen about its environmental safety: in 2007, the OSPAR marine protection treaty prohibited the storage of CO_2 in the sea water and on sea beds [83].

Assessment of Storage Potential

Estimates of worldwide storage capacity vary considerably, up to two orders of magnitude, and need to be consolidated by further research and studies as this is a key factor influencing the future scale of deployment for CCS and other sequestration options. While the potential in depleted oil and gas field can be reliably estimated, more uncertainty is related to deep saline formations, the largest storage resource. Recent academic literature has assessed that the global capacity is well above the extent of known fossil fuel reserves, by approximately one order of magnitude [87].

The International Energy Agency carried out in 2011 a very extensive study to assess the global storage potential [88], using as the main reference the assessment made by Hendricks [89], while for Europe estimations from [90] are used. For North America figures are updated using data from [91]. According to this review, global capacity is likely to be around 2000 Gt of capacity, with a best estimate around 11 000 $Gton_{CO_2}$: aquifers are the most widespread storage type worldwide, covering 85.6% of global capacity, followed by oil and gas fields (with 10.8%) and coal seams (with 3.6%).

4.3.1 Transport and Storage in TIAM

As it has been highlighted before, one of the advantage of Direct Air Capture is that it allows to mitigate decentralized emissions, not requiring to be located together with the source of emission. This flexibility of location could reduce transport cost, as capture is likely take place closer to storage sites, thus reducing the need for long pipeline to transport CO_2 from the capture facility to the sequestration site.

In the base version of TIAM, different commodities are defined to represent the stream of sequestered CO_2 coming from traditional CCS, further differentiating between industrial processes and fossil-based power plants, and from biomass-based plants with CCS (i.e. BECCS). The same commodity coming out from BECCS plants is used to characterize all technologies capturing carbon dioxide from the atmosphere, corresponding to Negative Emission Technologies. In this way, it is possible to track a range of sequestration strategies and assess their role in mitigation scenarios, modifying the relative costs and potential (see scheme in Figure 4.4).

These different flows of sequestered carbon dioxide are connected to the corresponding *sinking processes* representing distinct storage options: the list of different sequestration sites considered in the modeling can be found in Table 4.5. Moreover, these are further differentiated according to the origin of the CO_2 stream, whether it comes from traditional CCS in electricity or industrial sector, or from NET options (see scheme in Figure 4.4, where only the storage option related to EOR is represented.).

Transport costs are assigned to upstream *mining processes* for each storage option, that define also the constraint on the storage potential in each region. Differently, storage costs are assigned to the sinking processes.

Storage Options
Enhanced Coal Bed Methane < 1000m
Enhanced Coal Bed Methane > 1000m
Depleted gas field - OFFshore
Depleted gas field - ONshore
Depleted oil field - OFFshore
Depleted oil field - ONshore
Deep saline aquifer
Enhanced Oil Recovery

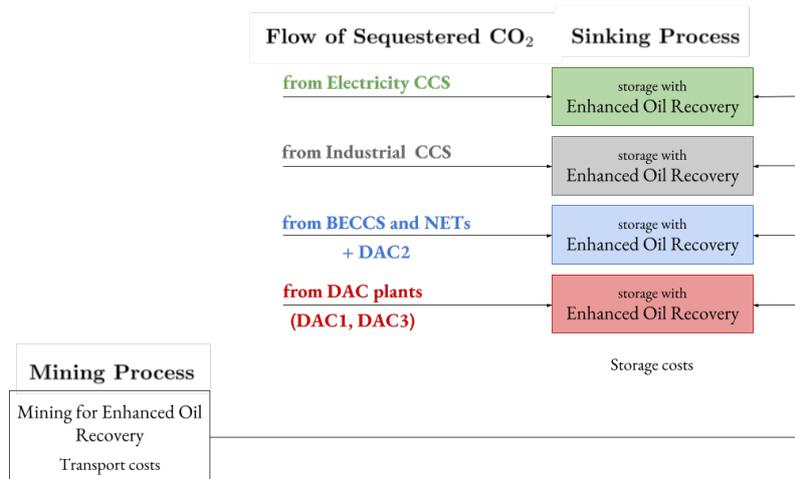Table 4.5: Storage options implemented in TIAM.

Figure 4.4: Scheme of storage and transport modeling within TIAM, referred to EOR sequestration.

In order to assess the impact of transport costs on the deployment of DAC, some modifications have been implemented in the TIAM structure, introducing a new commodity to keep track of the carbon dioxide captured only by DAC plants, independently by other CCS or NET options. This commodity is produced only by DAC processes and then sent to dedicated storage technologies: therefore, each group of storage and transport processes already defined in TIAM, has been duplicated to represent sequestration from Direct Air Capture plants. Initially, these have the same characteristics for cost and availability of the existing ones, but they will be modified to represent reduced transport costs for flexible DAC plants.

As the use of waste heat in amine-based DAC plants will constraint the location of these capture facilities, that need to be placed close to where heat is recovered, DAC2 processes are not characterized by the same location flexibility, thus they produce the same output commodity of other NETs.

Transport Cost

The transport cost currently considered in TIAM is 10 \$/ton of CO_2, which will be applied also to DAC sequestration in the base case[6].

In order to understand whether this aspect may influence the deployment of air capture, two cases have been considered for the transportation cost associated with these plants, reducing it to 5 and 1 \$/ton, without considering any differentiation across region. More refined analysis could be done by modifying the transport cost in each region and thus defining a sort of coefficient matrix to reduce/increase the cost per ton of CO_2 considering the average distance to different storage site: given that through diagnostic runs the influence of this parameter results quite limited, it has been decided not to go further in this direction.

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[6]Note that currently some regions are characterized by a lower or higher transport cost with respect to this baseline assumption, ranging between 3-10-30 \$/ton. Therefore, the same regional differentiation have been considered in the base case also for carbon dioxide coming from air capture facilities, so to have a homogeneous benchmark in my analysis.

Transport Cost	Base	Med	Low
[$/ton]	10	5	1

Cumulative Bound on Storage Potential

Estimates from IEA report [88] about cumulative potential are not so far from initial cumulative capacity implemented in TIAM (about 9500 Gt CO_2 available globally). Moreover, high, low and best estimates from Hendricks [89] are already available as storage potential scenarios in TIAM, with a global capacity equal to 5000, 1500 and 500 Gton respectively, then allocated to each sequestration option and each region[7]. These scenarios have been used for running a sensitivity analysis on storage capacity, and understand whether DAC and other sequestration options are in competition when the availability of storage is limited. As the analysis of many IAMs [92] showed that between 2010 and 2050 the storage demand will range between 100 and 500 Gt, when dealing with a 2°C target, capacity limits are expected to play a role only in the second half of the century.

Note that the constraint on storage availability has been defined as a cumulative capacity bound for each sequestration technology and each region. It has been decided not to make any intervention on how storage costs are defined within the model, as this is not the focus of my research work.

Global Storage Capacity	Initial	Hendriks, Low	Hendriks, Best	Hendriks, High
[Gt]	9400	555	1550	5030

4.4   Other Negative Emission Technologies

When dealing with Direct Air Capture, it is interesting to analyze the impact of this technology as part of a wider mitigation portfolio, and whether it is likely to be in competition with or to coexhist with other negative emission options, such as biomass-based power plants with CCS and afforestation. Indeed, it is unlikely that a single NET will be able to sustainably meet the rates of carbon removal consistent with 1.5°C of global warming [12]. If Negative Emission Technologies are to be deployed, a diversified portfolio could spread the risk across technologies, instead of focusing on one unique solution. For instance, DAC has the potential to reduce the impact needed to reach ambitious climate targets in term of land and water needs, though requiring larger amount of energy to work. In this perspective, it is important to understand which are the other sequestration technologies already implemented in TIAM and to assess carefully their cost and potential.

Beside traditional CCS applied both to coal-based and gas-based power plants, the technology options that are available in TIAM to remove CO_2 are:

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[7]Charts showing how the global potential is allocated to different regions can be found later in section 6.4, where results from different scenarios will be discussed.

- *Bio-Energy with Carbon Capture and Storage (BECCS)*, considering four different power plants and one technology producing hydrogen using biomass and electricity as input[8]. Electricity is produced both using dedicated crops, corresponding to the first generation of biofuels, and solid lignocellulosic biomass (i.e. second generation). The costs of these technologies have been updated in TIAM as they are the closest competitors for DAC (see next Section 4.4.2).

- *Afforestation*, increasing the amount of land covered by trees and forests that allows to capture and sequester carbon dioxide through natural photosynthesis. New technologies have been implemented in TIAM to represent this option (see Section 4.4.1).

- *Synthetic Fuel Production* from biomass, based on Fischer-Tropsch process and coupled with additional CCS unit, so to produce carbon neutral fuels For this technology, it was important to set an upper bound on its activity, as the model tends to over-install it due to the low input cost and the high value of synthetic fuel for transport sector (see Section 4.4.3).

Power Plants with CCS
Coal pre-combustion
Coal post-combustion
Coal Oxy-fuel
Gas post-combustion
Gas oxy-fuel

Negative	Emission Technologies
BECCS	biocrop with gasification
BECCS	biocrop with direct combustion
BECCS	solid biomass with gasification
BECCS	solid biomass with direct combustion
BECCS	H_2 production with CCS
FT synthesis	biocrop with CCS
Afforestation	carbon removal

4.4.1 Afforestation in TIAM

In order to model appropriately the afforestation option, two new technologies have been defined within TIAM to represent CO_2 removal, thanks to an increased plant stock through afforestation, and emissions, due to deforestation[9]. In order to define the amount of forests that can be installed, the different scenarios identified by the EMF21 (Energy Modelling Forum) are used as a reference in TIAM [93].

Within the EMF21, the role of carbon sequestration in forests was examined under a range of exogenously chosen carbon price paths, so to simulate several different climate change policies, ranging from 100 $ to more than 800 $ per ton of carbon by the end of the century, that means from 30 up to 220 $/ton$_{CO_2}$ (see table in Figure 4.5). Overall, this study shows that forestry is not an efficient measure for long-term policy alone,

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[8]This only plays a role when other NETs are switched off and DAC is very expensive, capturing around 2 Gt/yr: it is very costly but it consumes less electricity than DAC plants, 0.54 GJ/ton$_{CO_2}$.

[9]Note that previously this option was represented in the model with a lumped approach, with no possibility to install individual capacity unit of forests. As a consequence, it resulted an expensive option for the model and in most of the cases it did not enter in the solution at all.

but may represent instead an important long-term strategy to be combined with other mitigation options, such as DAC, due to its low costs[10]. According to other assessments [3], afforestation could deliver 1.1 up to 3.3 Gt_C/yr in 2100, corresponding to 4-12 Gt_{CO_2}/yr): EMF scenarios are consistent with these data, leading to a maximum of 1.8 Gt_C removed by afforestation in last decades of the century. Moreover, it should be noted the values of annual deployment resulting from EMF21 study are well in line with modeling results from recent reviews [12], being this an additional evidence that justify their adoption: review by Fuss [12] consider afforestation potential between 0.5 and 10 Gt/yr of CO_2 captured, while scenario 2 from EMF21 reaches a maximum amount around 7 Gt/yr).

Scenario	Initial Price	Growth
Scen 1	5 \$/ton$_C$	5 %/yr
Scen 2	10 \$/ton$_C$	5 %/yr
Scen 3	10 \$/ton$_C$	3 %/yr
Scen 4	20 \$/ton$_C$	3 %/yr
Scen 5	100 \$/ton$_C$	constant over time
Scen 6	75 \$/ton$_C$	+ 5 \$/yr up to 2050
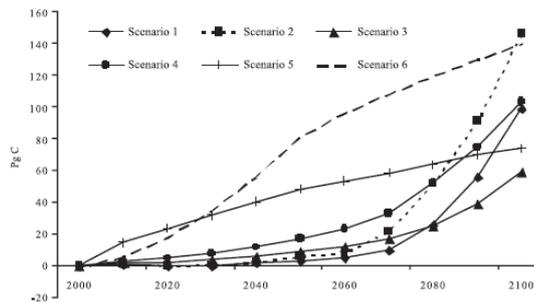
Figure 4.5: EMF21 afforestation scenarios: carbon price assumptions and cumulative sequestration.

Note that the different growth rates applied to carbon price (3 and 5%) in EMF scenarios reflect the different discount rate assumptions used in energy models: as in TIAM the discount rate applied is equal to 5% and we are dealing with stringent mitigation target, thus requiring higher carbon price, scenario 2 has been chosen as the most suitable reference to determine the afforestation potential in my analysis. In this case, carbon price scale rapidly up to 800 \$/ton$_C$ by the end of the century, that is about 200 \$/ton$_{CO_2}$: this value is lower than the marginal abatement cost usually reached in TIAM runs,it represents a likely cap for the carbon price to be applied in the agricultural sectors to avoid a negative competition with food, with respect to energy and industrial sectors where it needs to be much higher so to result effective. Note that the price for afforestation technologies defined in TIAM, including only operating costs, coincides with the carbon price applied in the corresponding EMF scenario.
Note that EMF study provides both the capture and the emission rate due to afforestation/deforestation mechanisms, even if only few countries have emission related to deforestation, namely China, India and Russia.

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[10]In this study, a dynamic global forestry model has been used, adapting the ones used in Sohngen and Mendelsohn (2003), that has been integrated also with DICE, part of the IAM family. The main limitation of the approach used is that it does not formally model agricultural markets.

4.4.2 BECCS: Cost Updates

BECCS represents an important mitigation option included by the majority of IAM-based scenarios aimed at keeping global warming below 2°C, as the ones included in the Fifth Assessment by IPCC (AR5) [2]: about half of the AR5 scenarios foresees BECCS exceeding 5% of primary energy supply.

As we have already discussed, TIAM results to be highly sensitive to costs when choosing the technology portfolio to be deployed, therefore it has been decided to update costs for BECCS and CCS plants and to align them to the ones used in other IAM studies. In particular, given that this research work may be further developed with an additional inter-model comparison using the WITCH model and considering that previous studies on DAC were developed using this model [4, 8], costs and efficiency for power plants with carbon capture have been aligned among the two IAMs, using a previous thesis work implementing CCS technologies in WITCH as a reference[11]. Differently, reference value for BECCS plants have been taken by the IEAGHG report [88], as it provides a differentiation between gasification and combustion plants (that is not present in WITCH model [94]).

Note that both initial and floor costs have been identified as well as high and low estimates for power plant efficiency, reflecting the energy penalty related to the capture process: an exogenous cost reduction rate equal to 4% has been implemented (see Section 4.1.4 for the discussion about technical learning rates), while efficiency improvements are considered to take place in the first half of the century, therefore the higher efficiency is reached by 2050.

Technology	CAPEX [$/kW]		OPEX [$/kW]		Efficiency [%]	
	start	floor	start	floor	start	floor
Coal pre-combustion	2740	1310	79	69	34	40
Coal post-combustion	2727	1310	104	69	34	39
Coal oxy-fuel	2896	1310	74	69	33	36
Gas post-combustion	1342	689	50	44	48	52
Gas oxy-fuel	1426	689	50	44	47	49
BECCS Gasification	2458	1826	77	56	28	38
BECCS Direct Combustion	3281	2566	93	62	28	33

Table 4.6: Cost and efficiency for CCS and BECCS. Note that the values are expressed in $_{2000}$.

4.4.3 Negative Emission Technologies Potential

When considering a diversified portfolio of NETs, it is important to assess which could be a feasible amount to be deployed along the century, together with potential economic, social and environmental implications related to these technologies. As TIAM does not incorporate land use or social aspects, the impact assessment of

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[11]Note that gas-based CCS plants are not considered in this work, therefore their cost is estimated considering that the same percentage difference between price of post-combustion and oxyfuel capture processes can be applied to both coal-based and gas-based plants

different NET options in term of land, water and material use will be done with *ex-post* calculations, as it will be discussed in next section 4.5. Moreover, it is important to define some constraints on the diffusion of these technologies in the model, as it has been already done for DAC alone.

According to the assessment by McLaren [78], DAC has the potential to reach level of 10 Gt_{CO_2}/yr, considering both supported amines and wet calcination, while BECCS will be able to capture between 2.4 and 10 Gt/yr by the end of the century.

According to [88], the overall economic potential for biodiesel production based on Fischer-Tropsch synthesis allows to capture around 3.3 Gt_{CO_2} by 2100. Considering recent studies [95], costs associated with BECCS capture are lower than for DAC case up to a removal of around 12 Gt per year, then they will increase abruptly due to biomass supply limitations.

An upper bound on the capacity installed have been identified for individual NET technologies as well as on the overall amount of CDR deployed, as it has already been discussed previously. Therefore, biomass-based FT process has been constraint to capture up to 3.3 Gt_{CO_2} in 2100, while afforestation results limited by the EMF scenario introduced before and BECCS by the availability of bioenergy (see next paragraph). The global potential for all NET processes has been set to be equal to 10 Gt CO_2 in 2050[12] and to 35 Gt by the end of the century as a base case, including DAC, BECCS, FT synthesis, hydrogen production from biomass and afforestation.

Bioenergy Potential

While DAC is assumed to be available at any level, BECCS potential is likely to be constraint by the amount of biomass that can be dedicated to power plants in a sustainable way, thus not entering in competition with food and water supply, as well as biodiversity conservation [14, 96]. Therefore, for this group of technologies it has been decided to put a limit not on the amount of CO_2 captured or the amount of electricity provided, but on the potential of bioenergy available, according to a range of model results analyzed by Searle [97].

Estimates on future supply of biomass are strongly influenced by assumptions on land availability beside nature conservation and food production, diet scenarios, land productivity and technological scenarios, energy crop yields and the supply of residues and wastes from other economic activities. After harmonizing a number of key assumptions[13] that lead to a very wide range of biomass availability, the study concluded that the maximum limit to sustainable energy crop production in 2050 will be between 40 and 110 EJ/yr, in terms of *Primary energy* from dedicated energy crops[14], with a median estimate of 80 EJ/yr. When also residues and waste are

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[12]This value has been increased from 5 to 10 Gton with respect to previous discussion on DAC alone, as according to EMF scenario 2 afforestation would take alone 3.7 Gton in 2050, thus leaving only limited space for other options.

[13]Linearity of final results with each input assumption was assumed, considering energy yields, available land, production cost, forest use, heating value and conversion efficiency

[14]Primary energy refers to inherent energy in the feedstocks before they are utilized.

considered, the potential estimate is projected to be between 60 and 120 EJ/yr, with 90 as median value. It should be noted that according to IEA accounting, bioenergy currently provides around50 EJ/yr on a global scale, that is about 9 % of global energy demand, most of it being represented by traditional biomass. Other assessments [98] conclude that above 200 EJ/yr sustainability issues are likely to occur.

In TIAM, upper bounds are already defined for each commodity representing different bioenergy source (waste, dedicated crops, solid biomass, biogas,...): in particular we are interested in dedicated biocrops (i.e. first generation of biomass) and solid biomass (i.e. second generation), as these are the ones fed in input to BECCS plants. The existing upper bound set for the dedicated crops is equal to 136 and 166 EJ/yr in 2050 and 2100 respectively, while for second generation is equal to 86 and 132 EJ/yr by mid and end of the century. The amount of bioenergy used in 2020 for baseline scenarios is about 63 EJ/yr. These values result to be much higher that sustainable estimates discussed previously, therefore it has been decided to update them by defining different scenarios for biomass availability so to perform sensitivity analyses on this parameter. Global biomass potential in 2050 has been set to be equal to 200 EJ/yr [98] as the base case, and then reduced to 120 and 90 EJ/yr [97], down to 63 EJ/yr, which is the amount employed in 2020 according to TIAM baseline scenarios. This cap in then kept constant up to 2100 and it accounts for all bioenergy, used both for electricity, biofuel production and heating, including also wastes and residues. Therefore, the constraint has been applied to the *mining processes* for bioenergy defined in the model. The allocation of this overall potential to individual regions is done using the same fractions already implemented in TIAM[15].

Currently in TIAM:

Bioenergy Mining Processes	Currently in TIAM		
	2005 (EJ/yr)	2050 (EJ/yr)	2100 (EJ/yr)
Industrial Waste	0.45	7.41	15.14
Municipal Waste	8.08	8.28	8.30
Biogas	0.52	7.50	
Liquid Biofuels	0.22	0.750	
1st generation Biocrops	8.56	136.1	166.0
2nd generation Biomass	45.52	86.48	132.0

Constraints Implemented in TIAM:

Global CDR Capacity	in 2100		
$[Gt/yr]$	Min	Base	Max
all NET	10	35	50

Global Biomass Potential	Low	Med	High	Base
$[EJ/yr]$ from 2050	63	90	120	50

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[15]As these coefficients change a bit from 2050 to 2100, it has been chosen to use the 2050 value.

4.5 Impact Assessment and Environmental Footprint

There are of course some limiting factors that need to be accounted when evaluating the potential of DAC: the success of this technology does not only depend on energy and costs, but also how these plants will impact on the environment, in terms of economic, biophysical and societal limits. While comparing different CDR options, one should consider the entire supply chain to evaluate their impact, in term of net emissions achieved, environmental footprint and resource use: this is not easy with IAMs, especially if a land model is not included as in TIAM.

According to the assessment made by Smith on physical and economic impact of large scale deployment of NETs [3], for Direct Air Capture technologies techno-economic requirements are expected to be the main issues that could slow its deployment, so that R&D should focus on reducing costs and energy inputs. Differently, BECCS may also be limited by nutrient demand and by a significant water use, both to irrigate feedstock and for the capture unit itself, eventually reflecting land constraints, while DAC may require much less land per ton of CDR than BECCS and may create fewer land-use conflicts.
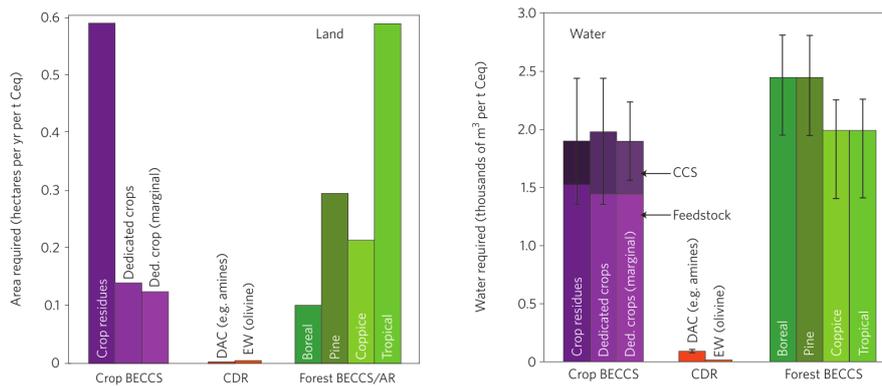
Figure 4.6: Impact assessment, from [3]

As a proper land model does not exist yet in TIAM, impact assessment will be done *ex post*, considering the need for water, land and materials (sorbent) to operate DAC plants. DAC will be compared with other NET deployment, as one of its advantages is the reduced footprint with respect to BECCS or afforestation. Differently, the major issue for direct capture is likely to come from the provision of NaOH for strong base reactions, as it involves high energy requirements and a change in the industry.

4.5.1 Land Use

DAC has minimal land requirements compared to BECCS that is fueled with biomass and crops. Direct Air Capture plants only require land for the construction of buildings: as there are only some pilot plants up to now, it is difficult to quantify the amount of land needed, also because different design proposed requires different structures. There is little risk for a buildup of CO_2 deficient air around the capture facility plant, since the atmosphere is effective at quickly and evenly mixing itself

[25], therefore the amount of land needed would be limited. Moreover, it can be deployed on unproductive land that supplies few ecosystem services. Note that the land footprint could increase considerably if solar PV panel or wind turbines were used to provide energy required.

According to [3], the land footprint related to BECCS power plants is between 270 and 1636 m^2 to capture 1 ton of CO_2 per year[16], according to the type of feedstock used to provide the input fuel: the lower end corresponds to purpose-grown crops, while more land is required for agricultural residues and even more when using forest residues to produce biomass due to their lower energy yield. In this latter case, the land use can go up to 1.7 ha/ton_C/yr, that is more than 4600 m^2/ton_{CO_2}, but it has not been considered within the range. Similar values can be applied also to afforestation, given that they are both biological sequestration strategies.

Moving to Direct Air Capture, the impact is much smaller, with amine-based plants requiring around 0.05-0.1 m^2/ton_{CO_2}/yr [41, 46] and the ones using hydroxide solutions around 1.5 m^2/ton_{CO2}/yr [20]. All these values are summarized in Table 4.7.

Land Use $[m^2/ton_{CO_2}/yr]$	Low	High
BECCS	273	1636
DAC	0.1	1.50
Afforestation	273	1636

Water Use $[ton_{H_2O}/ton_{CO_2}]$	Low	High
BECCS	545	682
DAC	5	20
Afforestation	545	682

Table 4.7: Land and Water Use for different NET technologies, from Smith, 2016

4.5.2 Water Use

Estimates of water required per ton of carbon removed by DAC is about one order of magnitude or more lower than for BECCS plants, demanding water both for growing crops and feedstock and for operating the CCS module. Again, similar values can be applied to afforestation [3].

Considering DAC systems, water loss represents a prime concern for some of them: aqueous systems are prone to evaporation, leading to a consumption of about 5 to 13 ton of water per each ton of carbon dioxide captured during normal operation, depending on humidity and temperature of ambient air [36, 99]. Differently, artificial trees may require up to 20 ton of H_2O [31]: indeed, this technology involves a dehydration step to release CO_2 after the moisture swing, requiring a significant amount of water. This may limit application of this technology to non arid regions. Developers of amine-based plants do not mention water use as a source of concern. All the values discussed here can be found in Table 4.7.

[16]They correspond to 0.1 and 0.6 hectares per ton_C

4.5.3 Material Use

Focusing on material use, besides the amount needed for the construction of the capture facility itself, it needs to be further considered whether the availability of sorbents proposed for DAC applications would be limited or whether it would result difficult to produce them when scaling up to large capacity.

Amine-modified sorbents developed by Climeworks and GT do not seem to have any risk associated. Considering strong base solutions, the impact on soil nutrient is almost absent and the only environmental risk may be associated with loss of hydroxide in liquid droplets into the atmosphere. Though, the production of these hydroxide sorbents is not straightforward, as large amount of energy is required, expecially to synthesize NaOH. Moreover, it should be considered that now this chemical is obtained as a side product from Cl_2 production, but in the future their respective roles may be reversed, with NaOH being the most valuable output once DAC reaches high level of deployment. Therefore it is important to explore the impact of DAC1 plants, and their need for sorbent makeup after each capture cycle, on NaOH production industry.

NaOH Production

Currently, sodium hydroxide NaOH - also known as lye or caustic soda - is obtained as a by-product from Cl_2 production process: electrolysis of concentrated sodium chloride solutions (brine) produces chlorine gas, hydrogen gas and aqueous sodium hydroxide.

$$2\,NaCl_{(aq)} + 2\,H_2O_{(l)} \rightarrow H_{2(g)} + Cl_{2(g)} + 2\,NaOH_{(aq)}$$
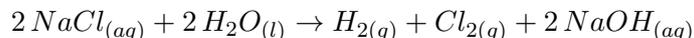
Being a high energy-intensive process (3.7 MWh/ton NaOH), it is important to consider also the impact in term of energy needed along the entire supply chain. Long-term changes in demand for NaOH will affect the least essential uses of NaOH, where it can be substituted by sodium carbonate (soda ash), such as in pulp and paper, water treatment, and certain chemical sectors where it is used as a neutralising agent[17]. According to APS report, there is a loss of sodium hydroxide solution during each capture cycle, as it remains partly entrained in the CO_2-depleted air leaving the absorber. Considering the detailed mass balance provided by Baciocchi [28], the make-up of sorbents needed is between 0.17 and 0.29 ton per each ton of CO_2 captured, in the different plant designs proposed.

KOH Production

Carbon Engineering is actually employing potassium hydroxide as a basic sorbent, differently from the APS benchmark: besides having more favorable binding properties, this material also lead to reduced environmental concerns. Nowadays, its production is based on electrolysis of potassium chloride solutions, which is analogous to the man-

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[17]According to: *Marianne Wesnaes and Bo Weidema, 2.0 LCA consultants, www.lca-net.com, 2006-10-19*

ufacture of sodium hydroxide, forming chlorine gas as a by-product:

$$2\,KCl + 2\,H_2O \rightarrow 2\,KOH + Cl_2 + H_2$$
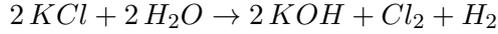
The main applications for potassium hydroxide (commercially called caustic potash) currently range from alkaline batteries to agrochemicals and fertilizers, food additives to soap and detergents, oil and gas drilling for the refining industry to de-icing fluids.

Currently the market for Chlorine Cl_2 is about 76.8 Mton/yr, that would allow about 300 Mton CO_2 captured using DAC1 plants. The current market for KOH is even smaller, about 0.8 Mton/yr, while about 100 times more NaOH is produced annually (about 80 Mt/yr). Therefore large scale deployment for this technology option would completely disrupt this market segment.

Material Use for DAC $[ton * /ton_{CO_2}]$	Low	High
NaOH	0.166	0.285
Cl_2	0.147	0.2666

Additional Energy $[GJ/tonCO_2]$	Low	High
for NaOH production	2.22	3.80

Table 4.8: Need for Sorbent Make up for DAC1 plants, based on Baciocchi analysis CITE, and additional energy required to produce hydroxide sorbents

4.6 Scenarios and Sensitivity Analysis

Generally in TIAM, energy demand levels are based on the socio-economic projections made in the Shared Socioeconomic Pathway 2 (SSP2) scenario [100], including population levels of peaking in 2070 at 9.4 billion before and an average annual economic growth of 3.13% from 2010 to 2050.

4.6.1 Mitigation Scenarios and Carbon Budget

Considering the current focus of the scientific community on stringent mitigation targets, different climate policy scenarios have been investigated, consistent with 2°C and 1.5°C increase in global temperature by the end of the century. In TIAM, this has been implemented by imposing a carbon budget, that corresponds to the amount of cumulative emissions allowed throughout the century to keep the temperature below certain limits. Indeed, it has been widely recognized the quasi-linear relationship between cumulative CO_2 emissions and temperature increase [1]. These budgets have been defined according to the reference commonly used in integrated modelling exercises [101], adjusting them to consider historical emissions up to 2015. Moreover, as TIAM does not account for industrial process emissions (e.g. cement industry), the budgets need to be further reduced: by doing so, the 1.5°C budget ends up being negative.

Mitigation Target	Carbon Budget 2016-2100 $[Gt_{cum}]$	Expected Industry Emissions 2016-2100 $[Gt_{cum}]$	Adjusted Carbon Budget 2016-2100 $[Gt_{cum}]$
2°C	810	233	577
1.5°C	220	233	-13

Table 4.9: Carbon budget defined in TIAM, consistent with a probability higher than 67% to keep the temperature increase below 2°C and 1.5°C respectively.

4.6.2 Summary of Sensitivity Analysis

According to experts [102, 16], there are some modeling challenges related to Carbon Dioxide Removal strategies and a number of key factors need to be addressed carefully. These include energy and cost requirements, and the related learning effects, possible diffusion pathways and physical scale that can be reached, the competition with other NETs, in particular BECCS and afforestation, also in term of storage availability and transport infrastructure. Moreover, inter-generational preferences should be taken into account, as IAMs discount future costs to different extents, therefore reducing the impact of future mitigation costs with respect to short-term ones.

Lot of uncertainty is still related to these key parameters, therefore the ranges highlighted during the previous discussion will be used to perform an extensive sensitivity analysis to check the robustness of results, both for 2°C and 1.5°C target. Each aspect summarized in Table 4.10 has been investigated independently, not to complicate the analysis too much, given the time and the tools available: more than 100 different scenarios have been examined, and the results will be discussed in Chapter 5 and 6.

Sensitivity Parameters		Levels Considered		
Energy and Cost Requirements	Energy Input	High		Low
	Cost Estimates	High		Low
	Learning Rates	15%	6%	1%
Diffusion	Annual Growth Rate	30%	20%	15-10%
	Maximum Capacity Allowed	50 Gt/yr	35 Gt/yr	20-10 Gt/yr
Storage and Transport	Storage Potential	Initial	High	Med/Low
	Transport Cost	10$/ton		1$/ton
Footprint and External Impact	Biomass Potential	200 EJ/yr	120 EJ/Yr	90-60 EJ/yr
	Impact Assessment	land	water	material
Intergenerational	Discount Rates	10%	5%	0%

Table 4.10: Parameters considered in the sensitivity, both for 2°C and 1.5°C scenarios.

Innovation with Previous IAM-base Assessment of DAC

Considering past modeling exercises including DAC in IAMs [4, 14, 5, 8, 6], this is the first work differentiating among different technology options to realize Direct Air Capture, while including references from demonstration plants to determine cost and energy estimates. Moreover, implementing the use of waste heat to be fed to these plants is another innovative aspect never investigated before. Considering the competition with other NETs, only [6] focused on the relative role of BECCS, DAC, afforestation and traditional CCS at the same time, but the environmental footprint was not considered, as well as the impact of a limited bioenergy availability.

Different storage availability have been already examined in [4], and [5], using Hendricks' estimates as a benchmark, but none of them tried to quantify the impact of a reduced transport costs due to location flexibility of DAC plants. Sensitivity on growth constraints have been investigated already in [4] and [8], as well as the impact of endogenous technical learning to model future cost reduction [4, 8, 5].

Moreover, this is the first attempt to develop and Expert Elicitation questionnaire regarding Direct Air Capture technology.

Chapter 5

The Role of DAC and other NET in 2°C-1.5°C Scenarios

In this chapter the results of my analysis will be presented, focusing on the impact of Direct Air Capture technologies as part of a wider mitigation portfolio with different climate targets. In particular, given the targets set by the Paris conference and the IPCC working groups, carbon budgets consistent with both 2°C and 1.5°C temperature increase by the end of the century have been taken into account. Indeed, more stringent mitigation targets would certainly require greater effort to decarbonize quickly the system and the possibility to achieve negative emissions is likely to have a larger impact. Before considering scenarios with a carbon budget, a number of diagnostic runs have been done, by applying a carbon price increasing in time, so to understand how the model would react to the availability of this new technology option and to check that everything was implemented correctly and was working as expected.

Along this chapter, it will be discussed the impact of the availability of DAC as a CDR option on the emission pathways and the energy sector (in Section 5.1 and 5.3), with a focus on its regional distribution (in Section 5.4). It will be further considered which is the impact of DAC on other mitigation strategies, whether they show complementarity or competition with the possibility to capture CO_2 directly from air (Section 5.6). Sensitivity analysis on different parameters will be discussed in next chapter.

Main Results: 2°C and 1.5°C Scenarios

Given the availability of DAC as a "backstop" technology, we want to investigate the impact on mitigation pathways meant to stay below 2°C and 1.5°C of temperature increase with respect to pre-industrial levels. For both climate policies, we are comparing different mitigation alternatives, so to understand what might change when trying to achieve ambitious climate targets relying only on BECCS and afforestation to achieve negative emissions (as it has been implemented in most of IAM-based studies so far, *No DAC* scenarios), or considering also DAC as part of the NET portfolio (*All NET* scenarios). In order to have a benchmark to evaluate the impact of *engineered* Negative Emission Technologies, it has been included also the case where neither DAC

or BECCS is available in the model (*No NET* scenarios). Note that afforestation is still included so to have realistic scenarios: indeed, without any possibility of carbon removal, model runs appear to be absolutely infeasible, with a carbon price exceeding 4 million $/ton and a mitigation cost 4 times higher than the projected GDP.

It should be noted that ambitious climate target, as the one consistent with a 1.5°C increase in temperature, cannot be achieved without relying on negative emission strategies: the carbon price reached in the *1.5C - No NET* scenario is still extremely high (more than 80 million $/$ton_{CO_2}$) even if allowing afforestation to capture 6-7 Gt/yr by the end of the century, confirming the urgency of further assessment and researches on CDR technologies. Nevertheless, this scenario has been included in the results shown in this section, but it does not represent a feasible and realistic mitigation pathway.

As it has been discussed in chapter 4, all these base scenarios allow a maximum CDR sequestration rate equal to 35 Gton/yr including also afforestation, and a bioenergy potential limited to 200 EJ/yr in 2100, considering updated costs for BECCS and CCS.
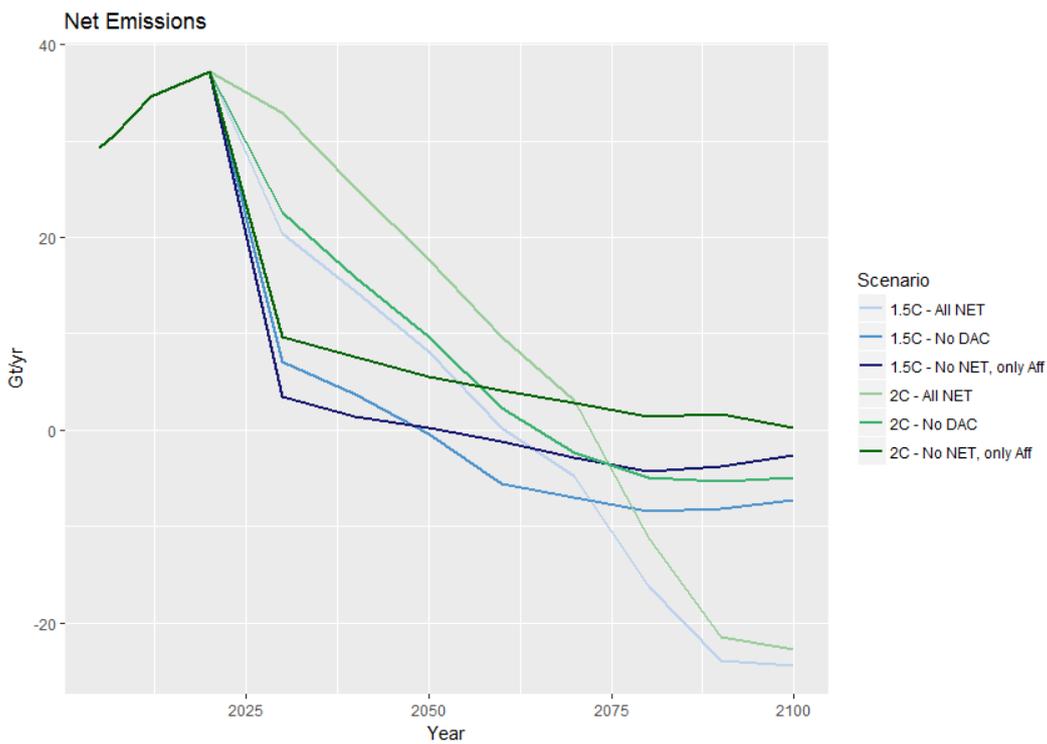
	Scenario	2020	2030	2040	2050	2060	2070	2080	2090	2100
2C	All NET	24	41	66	107	175	285	465	757	1233
2C	No DAC	24	117	191	310	506	824	1342	2186	3561
2C	No NET	24	4343	7074	11522	18769	30572	49799	81118	132132
1.5C	All NET	24	143	233	380	619	1008	1642	2675	4357
1.5C	No DAC	24	2119	3451	5621	9156	14915	24295	39573	64461
1.5C	No NET	24	2921408	4758666	7751365	12626157	20566679	33500952	54569521	88888000

Figure 5.1: Net emission pathways and carbon price in 2°C and 1.5°C scenarios.

5.1 Carbon Price and Net Emission Pathways

The possibility to remove carbon dioxide from the atmosphere will affect both the timing of mitigation effort, shifting the burden of emission reduction later in the century, and the level of net emissions reached by 2100.

As it can be seen from figure 5.1[1], without any CDR option (dark blue and dark green lines) a drastic decarbonization of the global economy is required in the first half of the century, with net emissions achieving slightly negative values by 2100 in a 1.5°C scenario (about -2 Gt/yr), while with a 2°C target the world will be almost carbon neutral. Both these scenarios foresee vigorous residual emission reduction rates in 2030, around 15-25% per year, especially in the energy sector. Indeed, here the role of CCS applied to power plants appear to be quite limited, as clean energy sources are adopted as the main mitigation strategy applied to power production, while CCS is largely adopted to decarbonize the industrial sector, with a capture rate up to 12 Gt/yr in 2°C consistent target. Residual emissions are about 11 Gt/yr and 7 Gt/yr 2°C and 1.5°C scenarios respectively.

It can be clearly seen that the availability of NET technologies, which are generally deployed in the second half of the century, reduces the effort in the near-term, requiring emission reduction rate smaller that 5% between 2030 and 2050 and shifting mitigation efforts towards the last decades with a less drastic trend. This is true when dealing with 2°C scenarios, while a more ambitious target still requires large decarbonization effort even when a full portfolio of NET is available (light blue line). In particular, it is interesting to notice that reaching a 1.5°C target relying on all NETs requires a similar emission pathways to the one consistent with 2°C without the presence of DAC between 2020 and 2070 (that is, before DAC capacity is being deployed to large scale, as it will be discussed in next section).

When DAC is available, net emission will be reduce significantly in the last decades of the century, with a rate lower than 1.5 Gt/yr due to the huge deployment of this technology that leads to a level of net emissions largely negative, around -24 Gt/yr by 2100 both in 2°C and 1.5°C scenarios. Indeed, Direct Air Capture is the main responsible for this strong carbon removal, hitting the overall constraint of 35 Gton removed each year by NETs, while BECCS and afforestation alone only capture around 17 Gt/yr, leading to net emissions close to -8 Gt/yr (*No DAC* scenarios). As a consequence, residual emissions are generally higher in scenarios with a full NET portfolio, as DAC allows the model to offset these additional CO_2 emissions later in the century: it should be remembered that the model operates with a perfect foresight.

Mitigation Costs

Besides influencing the emission pathways, it is interesting to notice that the availability of Direct Air Capture will reduce the overall mitigation cost to reach these ambitious climate targets, lowering significantly the marginal abatement cost (i.e. the

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[1]Note that all charts showing net emissions will include also the ones coming from cement production, that account for 2-3 Gt/yr by the end of the century.

carbon price) with respect to the case where only BECCS and afforestation can be used to remove carbon dioxide. With 2°C, DAC allows a 65% reduction in the value of carbon price, from 3500 to 1200 $/ton$_{CO_2}$ in 2100, while with 1.5°C it goes up to - 93%, going from more than 60 000 $/ton to about 4350 $/ton by the end of the century (see values in Figure 5.1).

When moving from a 2°C to a 1.5°C target, the difference is in the effort required already in the next decades: therefore, it results key from a policy-making perspective not to delay actions later in the century, as it is likely to increase dramatically the cost and the burden for next generations. The need for CDR technologies appears to be much more urgent with this stringent mitigation target, so to keep the carbon price and the total energy system costs within reasonable levels: indeed, the availability of a full portfolio of NET allows to achieve the same temperature increase with a more gentle emission reduction pathway (with rate around 1 Gt/yr) and a marginal abatement cost around 4000 $/ton.

Moreover, DAC also has an impact on the *energy system cost*: while with a 2°C target the availability of air capture technologies increase slightly the cost of the system (+7%) and consequently the mitigation cost[2], due to the higher expenses related with the deployment of this option both in term of capital and operational price, with a more stringent climate target the overall system cost is reduced of about 35%. Indeed, when trying to limit the temperature increase to 1.5°C, Direct Air Capture allows to avoid more expensive strategies in those sectors difficult to be decarbonised: looking at figure 5.3, it can be seen how CCS in the industrial sector plays a major role only when no DAC is available for the model. As a consequence, mitigation cost will account to around 4% of the projected GDP, rather than 9-10% when no DAC is deployed. Differently, in 2°C scenarios, the reduction in the carbon price thanks to the deployment of DAC within the NET portfolio (from 3500 to 1200$/ton) is partly offset by an increase in the overall cost of the energy system: being the carbon price more than halved, the 7% increase of the system cost suggests that the model is choosing to deploy relatively expensive DAC later in the century, raising the system cost in those years, but the present value (on which TIAM optimises) is still lower when there is DAC as those last decades are highly discounted.

It should be noticed than when DAC is available, the model treats it as a backstop technology, tending to over-installed it: without putting a cap its capacity would reach levels even higher than 90 Gt/year captured in 2100. Differently, BECCS capacity remains around 10 Gt/yr, as this technology is limited by the amount of bioenergy available. As explained before, DAC could be potentially deployed to any level given that there is no *explicit* bottleneck for the inputs needed for this process; I could not include any endogenous flow for the materials required for DAC within the model (e.g. amine and strong base sorbents), therefore the absence of external constraints makes

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[2]Note that the mitigation cost is defined as the difference between the energy system cost in mitigation scenarios with respect to a baseline scenario, where no climate policy is implemented and the main driver remains the economic and population growth.

DAC a preferred option for the model. Nevertheless, the impact of DAC in term of land footprint and water use will be evaluated later, in Section 5.5.

5.2 DAC Deployment and Cumulative CDR

DAC appears to be a long-term mitigation strategy across all scenarios, being installed at relevant scale only after 2070, while in the mid-term other strategies are more convenient to be deployed. This could also be related to the discount rate which is applied to future time periods, that result in a lower contribution of last decades costs in the overall objective function. This aspect will be investigated further with a sensitivity analysis (See Section 6.2).

Scenario	Technology	2070	2080	2090	2100
1.5C - All NET	DAC1	0.27	1.65	10.24	11.88
	DAC2	1.49	9.20	10.04	9.53
2C - All NET	DAC1	0.24	1.47	9.08	10.51
	DAC2	1.52	9.40	10.07	9.52

Figure 5.2: DAC deployment [Gt/yr] in 2°C and 1.5°C scenarios.

As already highlighted, the constraints of 35 Gt/yr of CDR is always hit when DAC is available, with this technology accounting for the majority of the overall potential: its capture rate is indeed around 20 Gt/yr out of 35 Gt/yr. The overall capacity of DAC plants installed and the allocation among different technology options is almost the same independently of the climate target imposed: in the 1.5°C case DAC removes only 1 Gt/yr more in the last decades than in the corresponding 2°C scenarios, meaning that the 20% growth constraint applied results binding in determining the extent reached by DAC deployment.

In both 2°C and 1.5°C scenarios, direct capture reaches a Gt scale in 2070, and by the end of the century the installed capacity is equally split between DAC1 and DAC2, each capturing around 10 Gt/yr. It does not result economically convenient

to produce heat on purpose to be fed to amine-based capture plants, as no DAC21 is being deployed. It is interesting to notice that plants based on amine-modified adsorbents are deployed earlier in time, while strong base solutions become a convenient option later in time: this may be related to the availability of natural gas commodity at lower price once it is not used any longer as a fuel in the power and industrial sector.

Looking at Figure 5.3, it can be noted that the role of CCS in the electricity sector is more relevant in the mid century, when other CDR options have not reached the maturity phase yet. It captures around 7-8 Gt/yr (in 2°C) peaking in 2060 and then declining in favor of BECCS and DAC, with a higher deployment in the scenarios with DAC, as additional electricity is required for this technology. With a 1.5°C target, the peak of CCS in the electricity sector is reached before, between 2040-2050, with a lower capacity installed globally, sequestering only around 5 Gt/yr.

It is interesting to notice that when other CDR options are not available at all, the deployment of traditional CCS is shifted prior in time, with 6 Gt/yr captured already in 2030, as a drastic and rapid decarbonisation of the electricity sector is required. After that time, its role flattens towards less than 1 Gt/yr captured in the last decades while sequestration from the industrial sector assumes a predominant role. In the *1.5C - No NET* scenario, a very small amount of carbon captured by traditional CCS can be found, as the huge decarbonisation occurs already between 2020 and 2030, reaching a level of net emissions around 5 Gt/yr and then becoming carbon neutral.
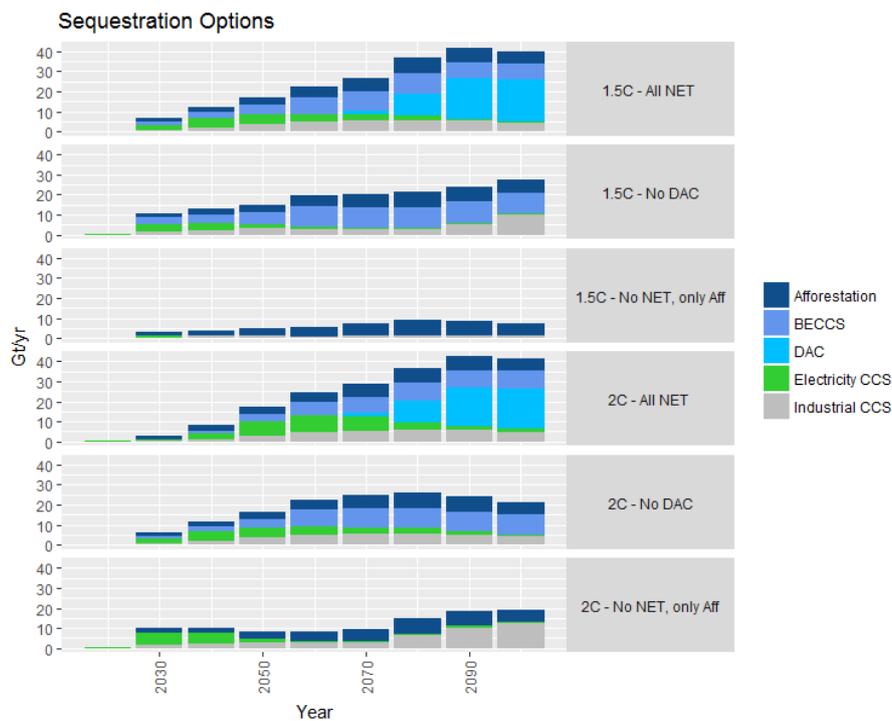
Figure 5.3: Deployment of other sequestration options [Gt/yr] in 2°C and 1.5°C scenarios, including BECCS, DAC, afforestation and traditional CCS.

When available, BECCS is deployed so to capture more than 8 Gt/yr in the second half of the century, from 2070 on: when DAC is not present, the deployment of bioenergy with CCS is shifted prior in time reaching 8 and 10 Gt/yr captured already in 2060 (for 2°C and 1.5 °C respectively) and keeping a stable capture rate around 10 Gt/yr for the last decades of the time horizon. Note that this level is consistent with other model scenarios assessing the sustainable potential for this technology option [?, 78]. Afforestation deployment results to be stable across scenarios, as it is limited by the level determined in EMF21.
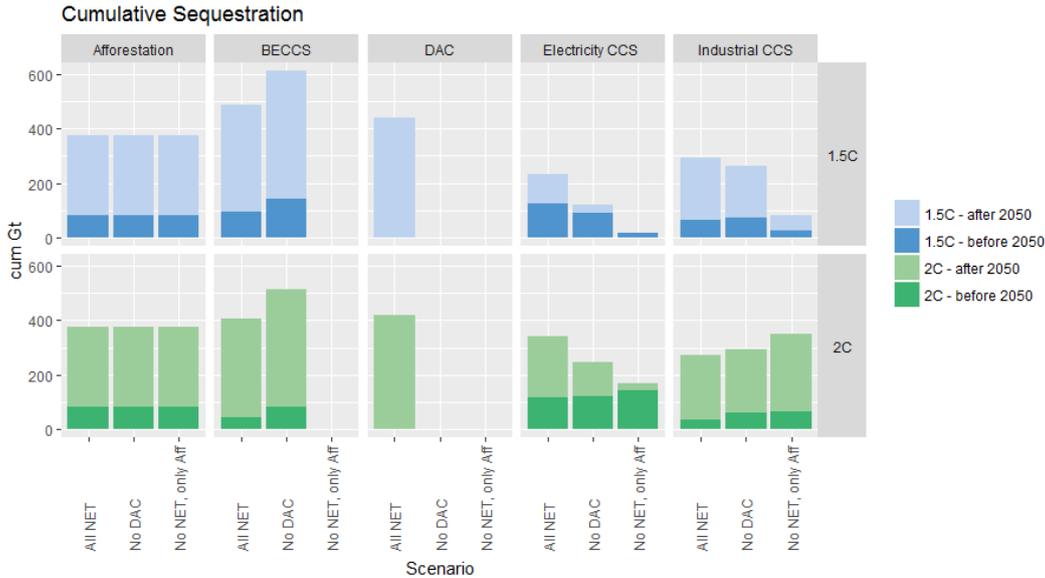
Figure 5.4: *Cumulative carbon capture along the century $[Gt_{cum}]$ in 2°C and 1.5°C scenarios, including BECCS, DAC, afforestation and traditional CCS.*

When investigating the different sequestration options available and their timing, it can be seen from Figure 5.4 that the availability of DAC increases the role of CCS in the electricity sector in the second half of the century, in order to provide the required energy to operate these plants with carbon-free sources. At the same time, carbon capture in the electricity sector appears to be reduced in the short term, as DAC allow to offset emissions from industry and transport sectors, requiring less drastic decarbonization in next decades. A huge impact can be seen also in the cumulative sequestration achieved by BECCS plants, both in the short and in the long term.

5.3 The Energy Sector

5.3.1 Electricity Production

Trying to reach 2°C mitigation target without relying on any technology for negative emission would have a marked impact on the energy system, requiring a huge deployment of renewable capacity already the coming decade (look at Figure 5.5).The general trend is that the availability of DAC as a NET option allows fossil fuel to still play a role in the electricity production later in the century, with coal still accounting for 20%

in 2050 in 2°C and gas for about 30% in 1.5°C scenario. This means that DAC may reduce the need for a drastic transition of the entire energy sector in the short term as it allows to offset emissions across all sector later in the century, being beneficial for those activities more difficult to decarbonize (look at sector emissions in next section). As the presence of only BECCS or only DAC as CDR options still requires massive transformation of the energy system, it can be said that there is an evident advantage in developing an integrated NET portfolio rather than focusing on one single option, and DAC should be carefully addressed when dealing with carbon removal technologies as it will play a fundamental role in the future to allow stringent climate target to be reached, reducing the global effort.

Of course, a stringent mitigation target (1.5°C) does not allow to still rely on coal for electricity production, nevertheless DAC allows less drastic transformation in the generation mix in the first half of the century, with an overall reduced demand for this commodity (-25%) that can be explained with a reduced need of electrification across different sectors. Moreover, the share of intermittent generation appears to be much smaller, from more than 75% to around 30%, containing the impact on the entire energy system in terms of costs and variability of supply throughout the year to be managed. Deploying the full NET portfolio allows to keep the share of intermittent renewables to around 50% by the end of the century in both 2°C and 1.5°C scenarios.
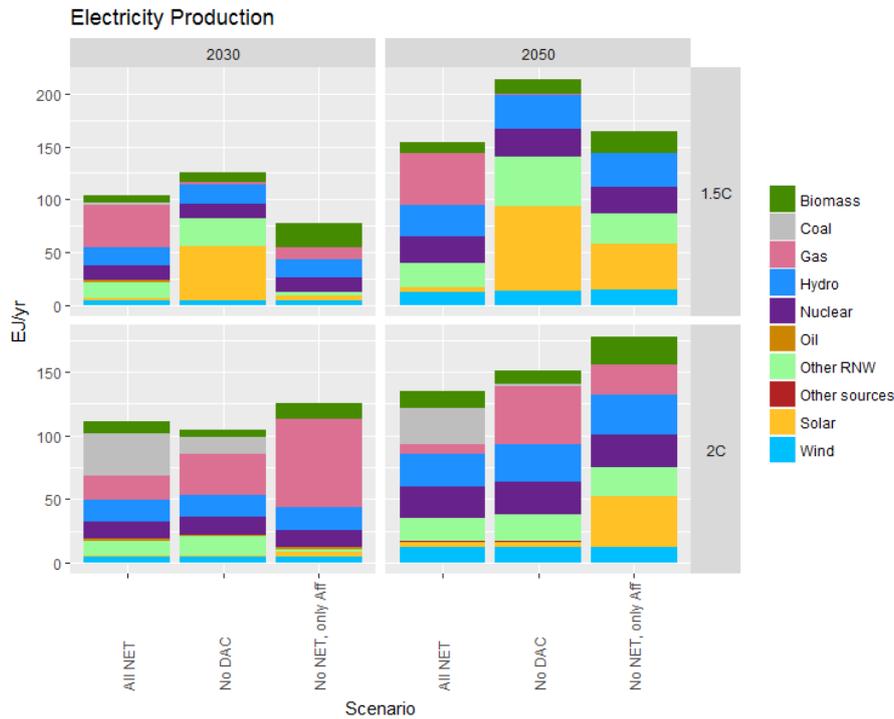
Figure 5.5: Electricity mix [EJ/yr] in 2°C and 1.5°C scenarios.

Looking at other scenarios both with 1.5 and 2°C target, it can be seen that the deployment of renewable capacity, and solar in particular, is higher when no DAC is available to meet the target, while deploying a full NET portfolio allows to install the lowest amount of renewables. As wind is tightly constrained to a 5% annual growth,

scenarios generally shows a huge deployment of nuclear, solar technologies (mainly PV, with more than 40 000 GW installed in 2°C) and hydro. Both PV and solar thermal show growth rates around 20% between 2020 and 2030, while other renewable, such as geothermal, tidal and wave, grow at an annual rate around 30% in the the first decade. These values represent the high end for a feasible growth scenario for clean technology, considering historical comparison, but they cannot be sustained for long time: indeed, such high rates usually apply only in the first development phase, after that maturity phase should be reached characterized by a more linear growth. As the growth of these technologies flattens to a 5% rate in next decades, it can be still considered a feasible pathways.

5.3.2   Total Primary Energy Supply - TPES

Similar impacts can be seen on the Total Primary Energy Supply (Figure 5.6), with direct capture technologies allowing to maintain a higher share of fossil fuel in the mix: coal and gas still hold a share of it along the century up to 2080 in 2°C and 1.5°C respectively. For 2°C, the availability of CDR options reduce the primary energy needed throughout the century with respect to the scenario where the same carbon budget is met only with low-carbon technologies, mainly due to the reduced role for renewable sources and biomass, but generally the presence of DAC will increase the TPES with respect to *No DAC* scenarios, both for 2°C and 1.5°C, given the large amount of energy needed to operate these plants. Indeed, in order to be operated, by the end of the century DAC plants require around 30 EJ/yr of electricity (accounting for less than 10% of overall production) and 160 EJ/yr of heat, both burning large amount of natural gas and by recovering waste heat.
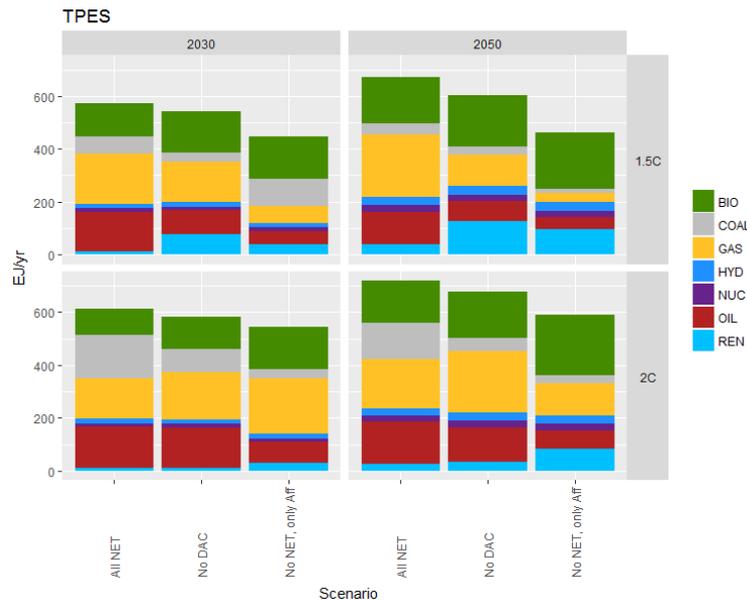
Figure 5.6: Total Primary Energy Supply [EJ/yr] in 2°C and 1.5°C scenarios.

Considering the price of commodities, the world without any CDR deployment is the one that shows the highest marginal cost for both electricity and heat, about 2-3 times higher for electricity and almost one order of magnitude higher when referring to heat commodity in 2°C scenario. The availability of DAC has an impact in increasing the cost of electricity and heat with respect to the scenarios without this capture option, as a greater amount of these commodities is needed to be fed into DAC plants, with both 2°C and 1.5°C.

5.3.3 Sector Emissions

As DAC allows to offset distributed emissions, without being linked to specific point source, is it interesting to notice from figure 5.7 that higher emissions are actually allowed from sectors such as transport and industry when a full portfolio of NET technologies is available, rather than when relying only on BECCS and afforestation. These corresponds to energy-intensive segments of the economy, difficult and expensive to be decarbonized in other ways. As expected, the level of residual emissions is the lowest when no CDR are deployed.

This aspect is even more evident in 1.5°C scenarios, where net emissions in the energy sector become negative already in 2070 in the *No DAC* case.
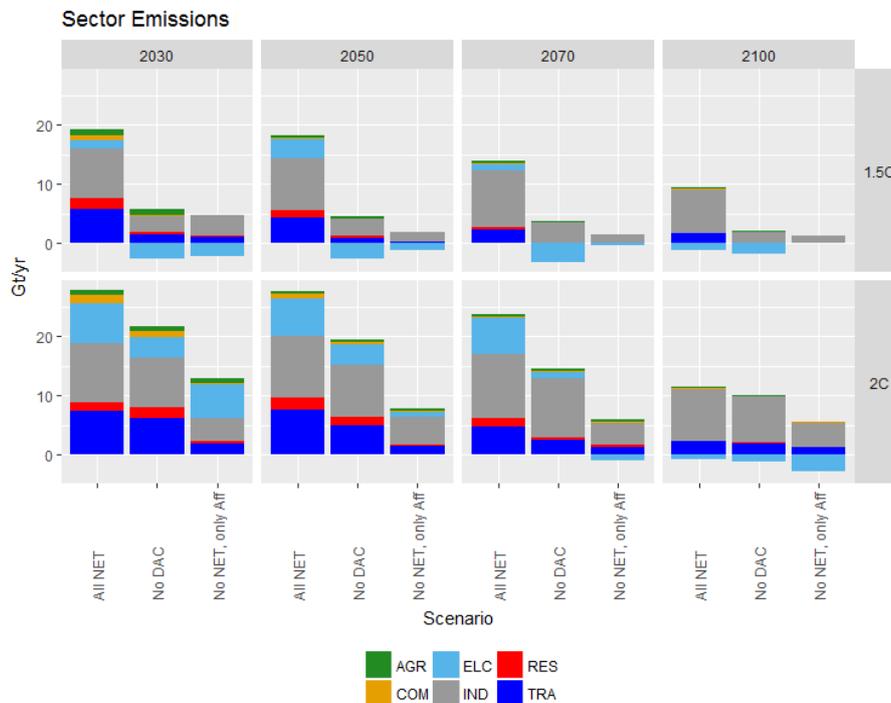
Figure 5.7: Sector emissions [Gt/yr] in 2°C and 1.5°C scenarios.

5.4   Regional Distribution

It results quite interesting to study the regional distribution of DAC together with other sequestration options, as it is shown in figure 5.8 for both 2°C and 1.5°C.

When available, DAC is deployed mainly in Western Europe (WEU), Asia (ODA - not China) and Central South America (CSA). While in Europe and Australia the predominant technology installed is DAC1, the one employing solutions of strong base, in Asia the installed capacity is almost half splitted between the two capture options, and in South America DAC2 accounts for the majority of the cumulative carbon dioxide captured. Some regions install almost only DAC2 (e.g USA, India, Africa and China), probably based on the large availability of waste heat.

When moving from a 2°C target to a more stringent one, it can be noticed that the cumulative capacity of DAC2 does not increase much, while more DAC1 is installed, mainly in Western Europe, Asia and Australia.
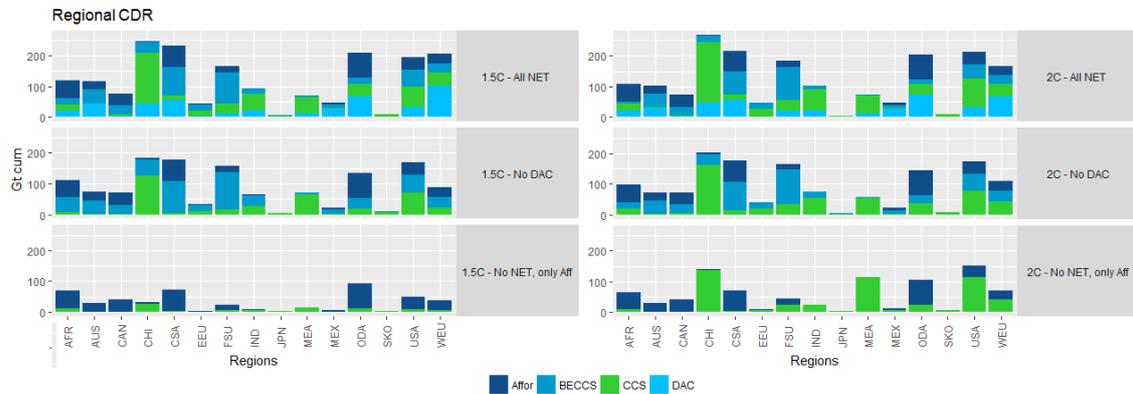
Figure 5.8: *Regional breakdown of different sequestration options. Cumulative capture along the century is considered [Gt_{cum}], in 2° C and 1.5° C scenarios.*

Considering the regional allocation all sequestration options (Figure 5.8), it can be noticed that China (CHI), Central South America (CSA), Asia (ODA), USA and Wester Europe (WEU) are the regions where the majority of emission offset takes place when all removal options are available to the model. CDR technologies are deployed mainly in central South America (CSA), Russian area (FSU) Asia (ODA) and Western Europe (WEU): while in the former ones this removal capacity is mainly represented by BECCS and afforestation, Asia and Europe rely greatly on installation of DAC plants. Differently, China is one of the regions where traditional CCS is still more relevant than negative emission options, equally split between carbon capture applied in the power and in the industrial sector.

Moreover, Middle East (MEA) tends to play a significant role in the case with no CDR technology, capturing more that 100 Gt cumulative along the century through traditional CCS, while with BECCS and DAC the role of this region is much reduced (to 60-70 Gt). At the same time, WEU starts playing a significant role on a global scale precisely thanks to the deployment of NET options, accounting for more than 150 Gt cumulatively captured, with a huge contribution of DAC itself.

It is interesting to notice that, even when a full portfolio of NET is available, there are still some regions whose net emissions are much higher than the amount that is removed from the atmosphere. This is particularly evident in China, India and to some extent also for USA. Regions like Australia, Canada and Mexico are able to bring their net emission almost to zero thanks to CDR options.

Generally, analyzing the competition between DAC and BECCS, it can be noticed that when air removal technologies become available, the capacity of biomass-based plants with carbon capture are reduced mainly in Africa (AFR), China (CHI) and Latin America (CSA).
When moving from 2°C to 1.5°scenarios, China and Western Europe strengthen their mitigation effort, increasing the installed capacity of traditional CCS in the first region and DAC in the second one. Differently, Russian area reduce the amount of DAC plants and USA reduce the amount of CCS: this can be explained with a reduction of their residual emissions to meet such a stringent climate target, thus requiring less CDR capacity.

5.5 Impact Assessment

When dealing with the role of CDR technologies to reach stringent mitigation targets, it is important also to consider the impact their deployment may have on other sustainable goals, mainly in terms of land footprint and water use.

Indeed, as previously discussed in Section 4.5, DAC has the advantage to reduce the **land** required with respect to a massive deployment of BECCS (see Figure 5.9a). Instead of requiring more than 26 million km^2 in 2100 (11% of global land for BECCS and 7% for afforestation) to reach the imposed climate targets, the deployment of DAC in the full portfolio can limit this to a 15%. It should be remembered that the availability of Direct Air Capture technologies reduces the role of BECCS of about 2-3 Gt/yr at the end of the century.
Exploring other combinations of NET options , it can be noticed that without afforestation the land needed to carbon removal technologies is reduced to 'only' 9% of global land[3], while relying only on DAC allows to further diminish it to almost 0% (0.05%). Of course this would mean higher mitigation costs for the system (i.e. higher carbon price and higher mitigation cost).

The same can be seen regarding **water** requirements: deploying also DAC in the NET portfolio allows to reduce the water needed of 5% and 15%, in 2°C and 1.5°C respectively, while relying only on DAC would require in 2100 only the 6% of the amount of water used when only natural removal strategies (i.e. BECCS and afforestation) are used. Therefore, combining DAC and other CDR options allows to reduce both land and water needs, of about the 8 and 5% respectively.

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[3]This mean that afforestation alone requires land for about the 7% of global ground surface.
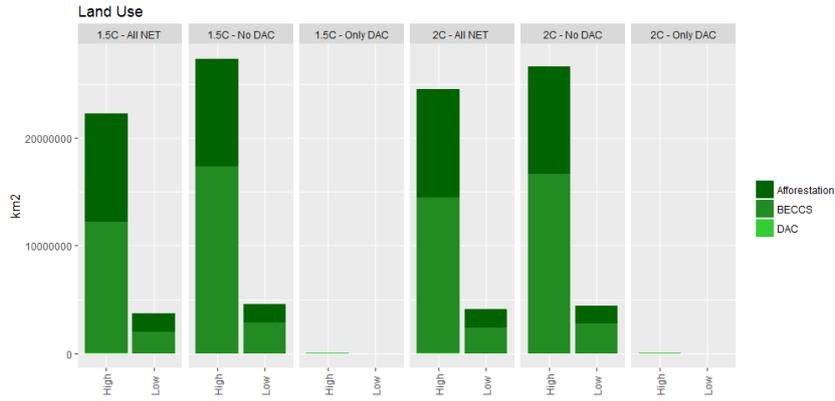
(a) Land Footprint [km^2]
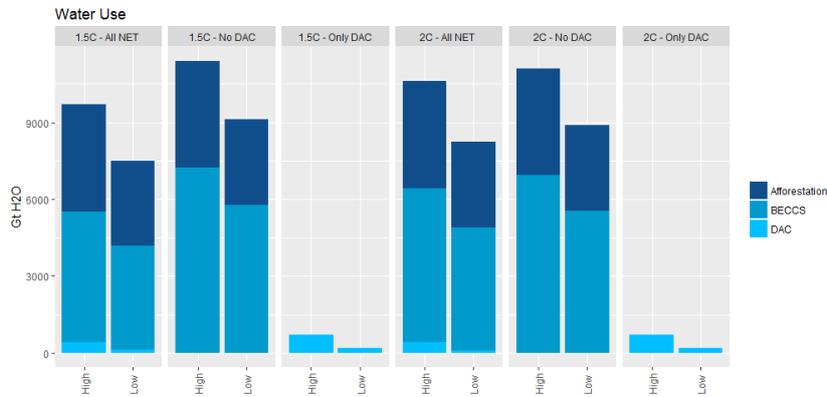
(b) Water Footprint [Mton$_{H_2O}$]

Figure 5.9: Impact Assessment, in term of land and water use, considering the level of deployment for different NET options in 2100, in 2°C and 1.5°C scenarios.

To fully assess the impact of DAC, it needs to be considered also the **materials** needed to run such plants. In particular, the strong base solution used in the APS design may bring some drawbacks related to its production process, as discussed previously. Indeed, the make-up of NaOH needed to capture around 10 to 20 Gt/yr with this kind of plants[4], would require around 3 to 6 Gt/yr in 2100 of sodium hydroxide, resulting in a production of chlorine of similar levels (3 to 6 Gt/yr as well, considering high estimate of make-up flow, while with a more efficient design this can be reduced to amounts between 1.5 and 3 Gt/yr). It should be remembered that currently NaOH is obtained as a by-product of chlorine, but chlorine demand today only accounts for 80 Mt/yr. This means that in the future the demand of NaOH will lead the production of Cl2, disrupting completely this market.

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[4]These are the levels of deployment for DAC1 plants foreseen by the model when Direct Air Capture is available together with other mitigation technologies, with and without afforestation respectively. When only DAC is available, all the DAC capacity installed is represented by this type of plants, that is 35 Gt/yr captured, but this extreme case has not been considered for the impact assessment.

As each ton of NaOH require 3.7 MWh to be produced (that means 3.80 to 2.22 GJ per each ton of CO_2 captured), the additional electricity required to operate such plants at this capture rate will be between 40 and 80 EJ/yr at the end of the century, which is a huge contribution considered the global level of production foreseen by the model, which is about 380-400 EJ in 2100.

Capture Rate	NaOH [Gt/yr]		Cl_2 [Gt/yr]		Electricity [EJ/yr]	
	Low	High	Low	High	Low	High
10 Gt/yr	1.70	2.85	1.50	2.70	22	38
20 Gt/yr	3.3	5.8	2.95	5.4	44	76

Table 5.1: Material use by DAC1 plants

5.6 Complementarity and Substitution with BECCS

In order to better understand the relative role of DAC and other Negative Emission Technologies, more scenarios have been analyzed, comparing different combinations of mitigation strategies: overall, I have considered the case with a full portfolio (*All NET*, with DAC, BECCS and afforestation), the ones with only the 'engineered strategies' without afforestation (*All NET - No Aff*), with only BECCS and afforestation (*No DAC*) and with only DAC deployed (*Only DAC*). The main focus was on understanding how the deployment of individual CDR technologies change when other competitors are available, as a limit on their overall capture capacity is fixed at 35 Gt/yr, or whether they may be considered as complementary to a certain extent.

The scenario showing the smallest carbon price at the end of the century is the one with a full CDR portfolio, both in 2°C and 1.5 °C. Differently, if DAC is used as the only option for achieving the mitigation target, the carbon price result much higher than employing only BECCS and afforestation: with 1.5°C increase in temperature, it results almost 3 times higher, reaching a value of 250 000 \$/ton, while with 2°C it is only 40% higher, confirming that this is quite an expensive technologies, though it may be beneficial as part of an integrated portfolio.

It is really interesting to notice that the deployment of other CDR options mainly have an impact on DAC1 technology: its installed capacity results to be mostly reduced in scenarios where DAC is coupled with BECCS, from more than 20 to around 10 Gt/yr in 2100, while capture plants based on amine sorbents are less affected (see Figure . For this technology, the plants using waste heat show an almost negligible change, while the ones consuming heat produced on purpose (i.e. DAC21) result not economically convenient any longer when a full portfolio of NET is available, while they are still able to capture around 2 Gt/yr in *Only DAC* scenarios with a 2°C target.
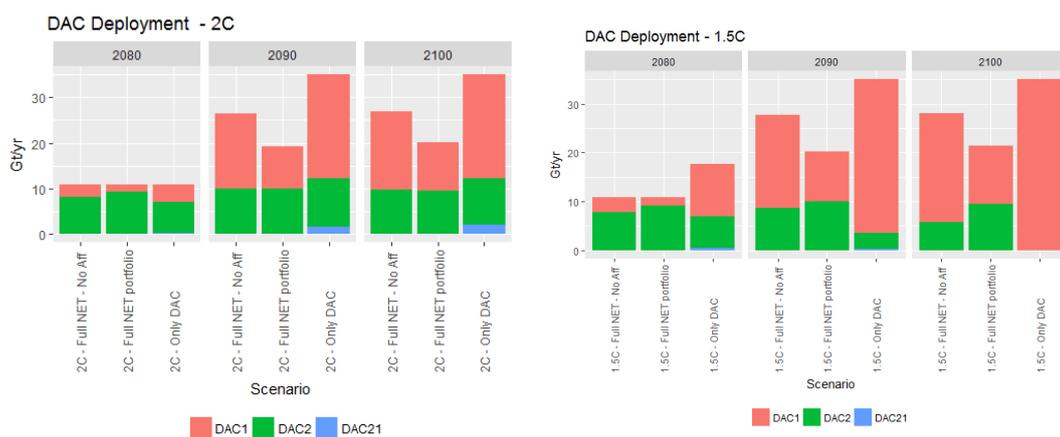
Figure 5.10: DAC deployment [Gt/yr], considering different mitigation strategies

When moving to a more stringent mitigation target, it is interesting to focus on the role played by different DAC technologies: while in 2°C scenarios the relative role of amine-based DAC (DAC2/21) is almost constant across scenarios with a capacity installed around 10-12 Gt/yr, with 1.5°C it is reduced to almost zero by the end of the century in the *Only DAC* scenario, with DAC1 accounting for the entire removal capacity allowed, that is 35 Gt/yr. It can be related to the fact that in *1.5C - Only DAC* scenario, when DAC1 is deployed so massively, the role of gas-based power plants for electricity production is almost zero already in 2050, while they still hold a relevant role in scenarios with other CDR availability. Therefore, natural gas is available in larger quantity to fuel DAC plants at cheap price. Differently, when also BECCS is available(*All NET* scenario), the role of DAC1 is reduced with respect to the case with only DAC and DAC2 is deployed up to 10 Gt/yr along the century: this shows a higher complementarity between the technology based on amine-modified solid sorbents and bioenergy plants with carbon capture. Therefore, it can be concluded that DAC1 is the technology being truly in competition with other NET options, as also its cumulative carbon capture is much reduced in scenarios where BECCS and afforestation are available.

Considering the **energy sector**, deploying only Direct Air Capture to achieve negative emissions, rather than as part of an integrated portfolio, has an impact mainly on the amount of electricity coming from solar PV, as solar thermal is able to provide also the waste heat needed by amine-based DAC plants. Moreover, in 2°C scenarios the phase-out from coal in the electricity sector takes place already before 2050, while in 1.5°C a larger share of electricity comes from solar already in 2050, with gas disappearing from the electricity mix as discussed previously. Nevertheless, the total amount of electricity needed in the last decades of the century is lower than in the case when no DAC is available, when dealing with a stringent mitigation target.

5.7 An Extreme Case

Artificial Tree and Floor Costs

During the literature review, three different technology options for realizing Direct Air Capture have been investigated (Chapter 2), but after the Expert Elicitation only two of these have been included in the main model scenarios. Indeed, experts tend not to consider the Artificial Tree proposed by Lackner a feasible option, as few data have been provided so far and no demonstration plant has been built yet (see Chapter 3). Nevertheless, I was interested in understanding the possible impact of this technology if it would be actually realized at a commercial scale, given that it potentially shows advantages with respect to the others, requiring only electricity as an input.

Therefore, additional scenarios have been investigated, considering that the full portfolio of DAC technologies is supposed to be available in the future: in the *Extreme scenario* considered in this section, in addition to the introduction of DAC3 in the model, floor costs are applied to all DAC technologies starting immediately from 2030. Moreover, captured CO_2 can be used to produce synthetic fuel (methane), given that some companies are planning to do it with their first plants, as it has been discussed in previous section 2.8: even if this process is allowed within TIAM model, it is never deployed during these runs, due to the high costs associated with it compared to the other paths available to produce methanol.

Scenario	Technology	2070	2080	2090	2100
1.5C - All NET	DAC1	0.27	1.65	10.24	11.88
	DAC2	1.49	9.20	10.04	9.53
	DAC3	-	-	-	-
1.5C - All NET - Extreme	DAC1	0.20	1.22	7.58	9.15
	DAC2	1.55	9.63	11.24	10.97
	DAC3	0.00	0.00	2.00	2.14
1.5C - All NET - Extreme3	DAC1	0.00	0.00	0.00	0.00
	DAC2	1.06	6.60	10.00	8.93
	DAC3	0.69	4.25	11.38	14.64
2C - All NET	DAC1	0.24	1.47	9.08	10.51
	DAC2	1.52	9.40	10.07	9.52
	DAC3	-	-	-	-
2C - All NET - Extreme	DAC1	0.14	0.89	5.51	7.25
	DAC2	1.61	9.96	12.58	11.72
	DAC3	0.00	0.00	1.61	2.25
2C - All NET - Extreme3	DAC1	0.00	0.00	0.00	0.00
	DAC2	1.44	8.77	9.99	9.09
	DAC3	0.31	2.08	10.46	13.76

Figure 5.11: DAC deployment [Gt/yr] in Extreme scenarios, including DAC3 and floor cost estimates.

The introduction of the third DAC option does not have an impact on the overall emission path and on the carbon price, neither on how traditional Carbon Capture and Storage is being deployed in other sectors along the century. However, it does influence the overall system cost, which results at the end of the century 8-10% lower than scenarios without the Artificial Tree.

DAC plants based on Lackner design only have a limited role in the last decades (up to 2 Gt/yr captured) when high cost estimates are applied to DAC3 (*Full NET - Extreme* scenario), while when all technologies are assigned floor cost estimates (*Full NET - Extreme Floor3* scenarios), it is going to take most of DAC capture capacity, sequestering around 13-14 Gt/yr out of 35 Gt/yr (see Figure 5.11). Indeed, for DAC3 the capture cost result to be around 30 $/ton, while DAC2 and DAC1 require 50 and 100 $/ton respectively. It is interesting to notice that even in these extreme cases when floor cost estimates are applied already in 2030, the deployment of air capture plants does not start much earlier in time compared to other cost scenarios. Once again, the growth constraint appears to be binding for the model, representing the prevailing aspect in determining the timing of DAC diffusion. An extensive sensitivity analysis on the impact of this growth constraint on model appear to be essential, and it will be discussed later in Section 6.3.

(a) Energy Input to DAC technologies
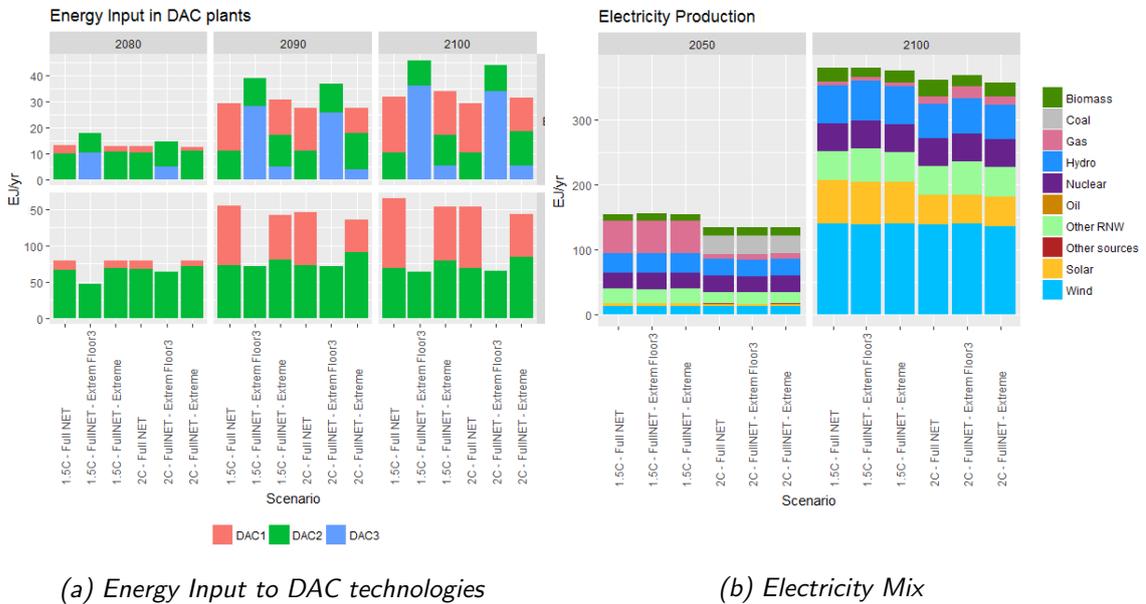
(b) Electricity Mix

Figure 5.12: Energy required by DAC technologies and Electricity mix in Extreme scenarios

While the impact on the rest of the system is limited, it is interesting to note how the choice among DAC technologies is influenced by the availability of a third option. Indeed, looking at the bar chart in figure 5.11, it can be seen that DAC3 is in competition mainly with DAC1, as these plants are not deployed any longer once artificial trees are available, while it shows quite a good complementarity with DAC2, whose deployment does not result much affected across scenarios. Indeed, part of the amount of gas that is not dedicated to DAC1 power plants any longer can be used in the power sector to produce the electricity needed by DAC3 plants, as it can be noticed looking at the shares in the power sector from figure 5.12b.

Even if the electricity demand for DAC power plants increases by 30-50% due to the fact that DAC3 requires almost twice as much electricity input compared to the other technologies (see Figure 5.12a), there is not a huge impact on the electricity mix and the global demand for electricity: the overall increase is quite limited, leading to a maximum of 20 EJ/yr more than the base case.

More solar PV capacity is actually installed when a large amount of DAC3 is installed, corresponding to the scenario when also this technology has floor costs, in order to face the increased demand for electricity required by artificial trees, while more solar thermal capacity is needed in the scenario where floor costs are applied only to DAC1 and DAC2, corresponding to a larger amount of amine-based plants requiring waste heat as an input (Figure 5.13).
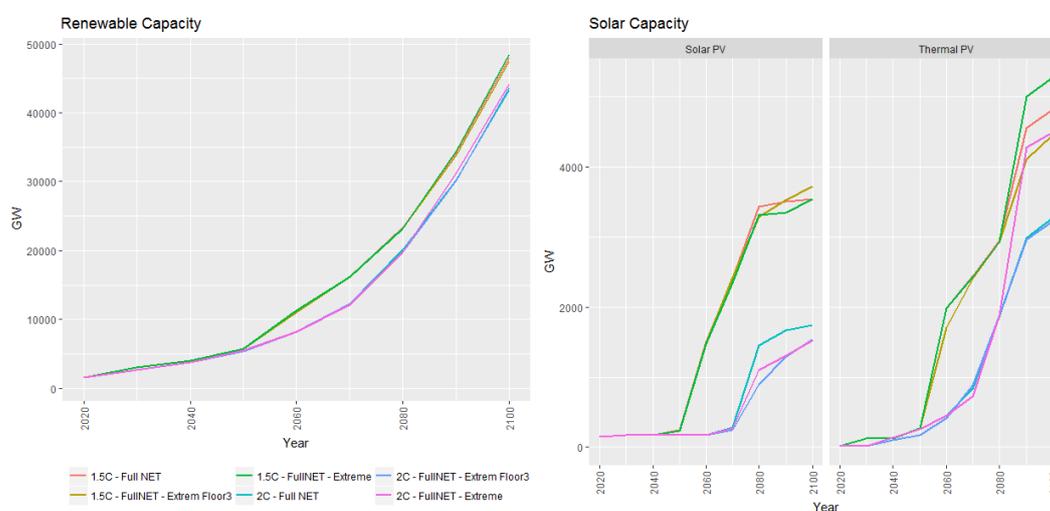
Figure 5.13: Renewable capacity installed [GW] in Extreme scenarios.

Chapter 6

Robustness Analysis

As previously discussed, there is a number of uncertainties inherent in estimating benefits and costs of mitigating climate change, given that each input parameter used in our Integrated Assessment Model is uncertain in the long run. Moreover, it is almost impossible to forecast which low-carbon technology will be invented in the future and how the costs of current technologies will evolve, as disruptive and unforeseen changes are likely to take place in time, as it happened for the rapid cost reduction and consequent diffusion of solar PV in the last decade. Therefore, in order to check the robustness of my results, sensitivity analysis has been carried on the main parameters defined to model Direct Air Capture technologies in TIAM, to understand how much their variation may influence the deployment of DAC along the century.

In particular, it has been investigated the impact of different cost and energy input estimates (Section 6.1), the inter-generational time preference (i.e. the time discount rate, Section 6.2), the various growth constraints applied, both in term of annual growth and maximum capacity (Section 6.3). In addition to that, different assumptions on storage availability (Section 6.4) and biomass potential (Section 6.5) have been defined in the model, to investigate further the interaction between DAC and other NET options.

6.1 Sensitivity on Energy and Cost Assumptions

As there is still lot of uncertainty about the energy requirements and the costs related to Direct Air Capture technologies, being it in the early stage of development, it is useful to run some sensitivities on these assumptions, and understand how much the cost estimates and the energy needs impact on the deployment of DAC.

In particular, different scenarios have been considered, referring to the values identified in Section 4.1.1 and 4.1.3, both with 2°C and 1.5°C targets: the *BASE* case is with high cost and energy estimates, then the effect of low cost and low energy input are analyzed separately in *Low Cost* and *Low Energy* scenario respectively. In addition to that, an exogenous cost reduction at 6% per year is considered, starting both from high and low estimates (*High Exog Reduction* and *Low Exog Reduction*), and this is also investigated in combination with improved energy performances, given

the peculiarity of this scenario (*Low Energy - Low Exog Red*). Moreover, the impact of very high cost has been included as well (*Very High Cost*), applying a capture cost equal to 600\$/ton for both DAC1 and DAC2, according to the common reference price applied to DAC[1] [20].

Scenarios Investigated:

Scenario	Cost	Energy	Cost Reduction
BASE	High	High	No
Low Cost	Low	High	No
Low Energy	High	Low	No
High Exog Reduction	High	High	6 %/yr
Low Exog Reduction	Low	High	6 %/yr
Low Energy - Low Exog Red	Low	Low	6 %/yr
Very High Cost	600 \$/ton	High	No
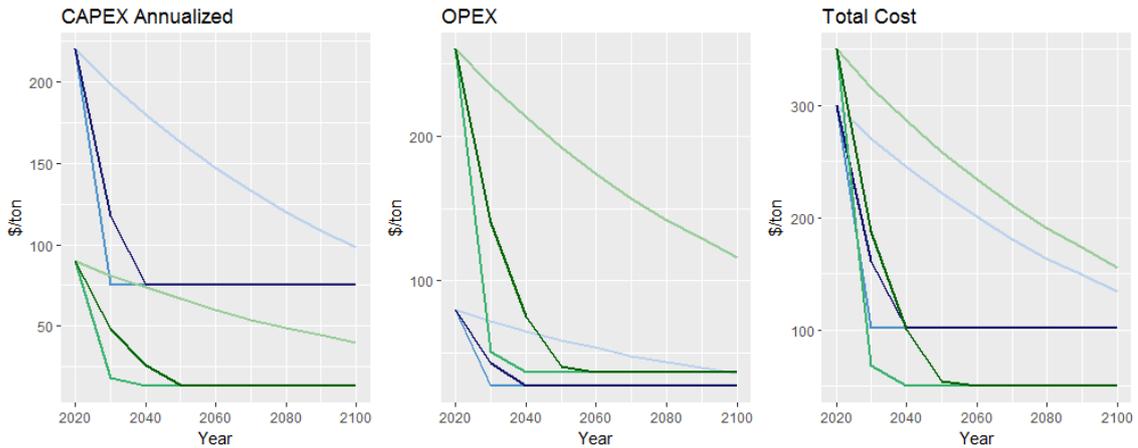
Figure 6.1: Exogenous cost reduction profiles, applying different reduction rates

Besides these scenarios, also the impact of *Low Transport cost* have been explored, combining it with different cost and energy estimates. Across all scenarios it has been seen that the impact of this parameter is almost negligible for the model, given the importance of deploying DAC to reach stringent mitigation target. Therefore, these scenarios have not been included in the following discussion.

As it can be seen from figure 6.1, different cost reduction rates can be applied to model technical learning, resulting in different cost profiles in time. It should be noted that applying a 1% annual reduction the floor cost is not even reached by the end of the century, while with 15% it is realized immediately, in 2030, that is much before the real deployment of DAC. The impact of various cost reduction rate will be discussed briefly later.

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[1]Note that this overall capture cost is then split between capital and operational expenditure differently for each DAC technology, according to their peculiarity discussed previously.

6.1.1 2°C Scenarios

Growth constraint applied

The results which are going to be discussed in this section are characterized by an imposed 20% annual growth rate, so that the impact of energy and cost assumptions appears to be very limited: indeed, this constraint results quite binding in determining the level of DAC deployed each year up to 2080. In the last decades a certain degree of variability can be found across scenarios. On the other hand, it cannot be avoided, otherwise the model would over-install this technology, with infeasible diffusion rates.

Net Emission Pathways and Carbon Price

As it has been said before, the impact on the net emission pathways is almost negligible as the growth constraint mainly determines the large scale installation of NETs. Compared to the base case, the carbon price results lower in all scenarios, but in most of them the change is less than 1%: with *Low Cost* it is almost the same, which would not be expected. The scenarios with a more marked impact are the ones with *Low Energy* estimates, showing the smallest carbon price around 1000 $/ton when considered alone and around 1100 $/ton when they are combined with a cost reduction, resulting in a 15% and 10% reduction with respect to the base scenario respectively.
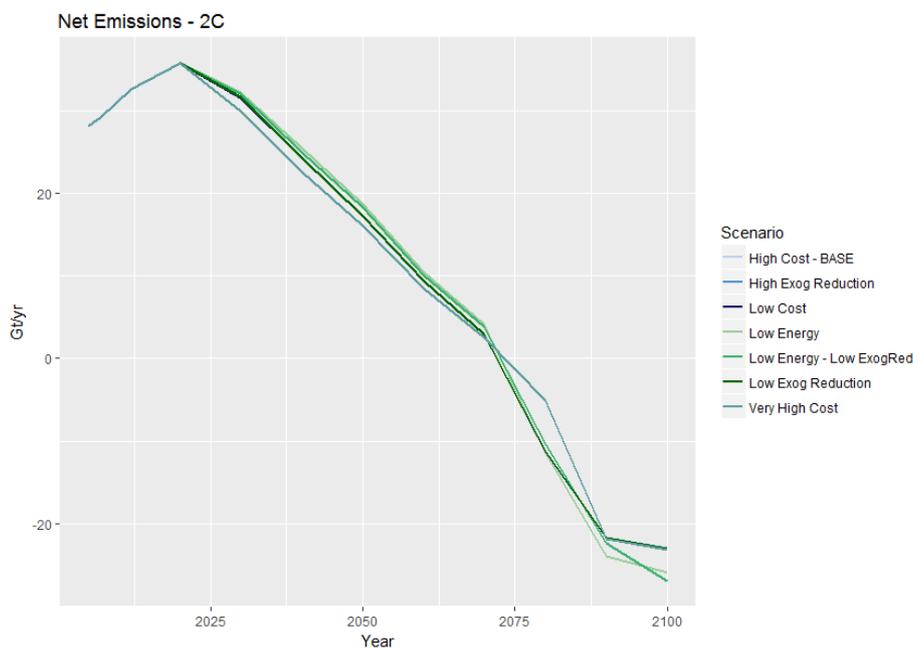
2°C Scenario	2020	2030	2040	2050	2060	2070	2080	2090	2100	%
BASE - High Cost	24	41	66	107	175	285	465	757	1233	
High Exog Reduction	24	40	66	107	174	284	462	753	1227	-0.5%
Low Cost	24	40	66	107	175	285	464	756	1232	-0.1%
Low Energy	24	35	56	92	149	243	397	646	1052	-15%
Low Energy - Low Exog Reduction	24	37	60	97	158	258	419	683	1113	-10%
Low Exog Reduction	24	40	66	107	174	284	462	753	1227	-0.5%
Very High Cost	24	50	81	132	215	351	572	931	1516	+23%

Figure 6.2: Net emission pathways and carbon price for 2°C, with different cost/energy assumptions.

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Looking at the total system cost, it can be noticed that cost and energy assumptions have a different impact: indeed, in the *Low Energy* scenario the system cost is almost the same as in the base case, while it is being reduced of 5% on average in scenarios with reduced cost assumptions. This is different from the impact that cost and energy has on DAC deployment, which results to be more influenced by energy assumption rather than cost as it will be discussed later.

DAC Deployment

From 2080 on the different scenarios start differentiating from each other, not only in the deployment of individual technologies but also in the overall diffusion of DAC with respect to other NETs, especially BECCS as afforestation is almost fixed.

In particular, the scenario combining low energy estimates and exogenous cost reduction (*Low Energy - Low Exog Red*) is the one that shows the highest deployment of DAC, up to 31 Gt/yr in 2100 out of the 35 Gt/yr allowed. Note that capture by afforestation is not present in the last decade as DAC results more economically convenient, while at the same time this scenario shows the smallest deployment of BECCS, whose role is reduced from 2080 on, down to less than 4 Gt/yr captured in 2100 (see Figure 6.4). As expected, reducing separately the costs throughout the century and the energy input required increases the amount of DAC deployed of more than 1 Gt/yr in 2100 (*Low Exog Red* and *Low Energy*), but the combined effect of these parameters is much higher than the sum of the two scenarios.

Considering the different technology options (see Figure 6.3), DAC21 is not deployed at all as we are analyzing scenarios where DAC compete with other NET options: indeed, we have shown previously that it plays a role only when other CDR are not available to the model besides DAC, otherwise it results more convenient to exploit waste heat for running amine-based plants.
With baseline assumptions (that is, high cost and high energy estimates) the installed DAC capacity is almost equally split between DAC1 and DAC2 in the last two decades of the century[2]. Reducing the cost exogenously during the century, DAC2 results favored on DAC1, increasing its capacity to 13 Gt/yr captured, while only around 7 Gt/yr of DAC1 are being installed: this means that around 30% more amine-based capture plants are deployed. It should be noted that applying an exogenous reduction, the floor cost reached for DAC2/21 is lower than for DAC1, as it can be seen from figure 6.1. Moreover, the difference between scenarios where exogenous cost reduction starts from high or low cost estimates is almost negligible, as to demonstrate that what really matters for the model is the value of the floor cost reached in the last decades of the century representing the period when DAC is actually deployed

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[2]It should be noted that up to 2080 the amount of DAC2 is much higher than DAC1, with 9 Gt/yr captured versus 1Gt/yr, as the former technology is more convenient to be deployed earlier in the century.
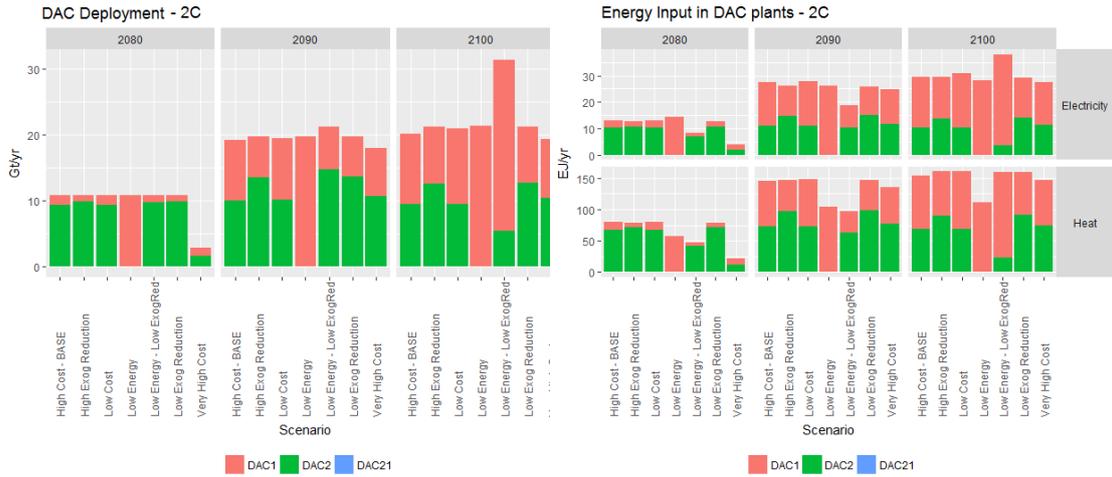
Figure 6.3: DAC deployment [Gt/yr] and energy required to operate it [EJ/yr] for 2°C scenarios, with different cost/energy estimates

2°C Scenario	2070	2080	2090	2100
BASE - High Cost	1.75	10.85	19.15	20.04
High Exog Reduction	1.75	10.85	19.70	21.21
Low Cost	1.75	10.85	19.45	20.86
Low Energy - Low Exog Reduction	1.75	10.85	21.20	31.31
Low Energy	1.75	10.85	19.80	21.28
Low Exog Reduction	1.75	10.85	19.70	21.21
Very High Cost	0.47	2.90	17.94	19.26

Differently, when reducing the energy need, DAC1 results the most favoured, as the model decided to install exclusively this technology: already in 2080 the capacity installed of DAC1 is about 10 Gt/yr (compared to the base case where it was only 1 Gt/yr), then reaching more than 20 Gt/yr captured by 2100. Note that the energy estimates for DAC1 in the *Low Energy scenario* are still higher than the ones for DAC2, nevertheless the model prefers to install exclusively this technology as probably the levelized cost of capture results more convenient. Indeed, DAC2 has higher operational costs than DAC1 due to degradation and stability issues related to the sorbent, so that the energy input cost will represent only a small fraction of the overall cost of capture, therefore its reduction does not favor this technology so much, considering also that waste heat comes almost for free.

The most interesting scenario results the one where low energy estimates are combined with an exogenous cost reduction: indeed, these factors individually would favor one of the two DAC technology option, DAC1 and DAC2 respectively. Combining them, it result that DAC capacity increases of 50% at the end of the century: in particular, DAC1 is the most favored, as it almost doubles its capture rate with respect to the baseline, while also reducing the role of BECCS in the last decades. On the other hand, DAC2 is deployed more up to 2090 but then it is much reduced in the very last decade due to the high amount of DAC1 installed. Remember that this is the last period of our time horizon, so that it results highly discounted.

Even in the case with extremely high cost [3], the amount of DAC deployed does not change much at the end of the century, with around 1 Gt/yr less captured in 2090 and 2100 decades and DAC2 slightly favored rather than DAC1, but its deployment starts massively later in time, after 2080, when the overall system cost is heavily discounted in the objective function. Therefore, we could conclude that Direct Air Capture technologies appear to be cost effective across deep mitigation scenarios, even when high costs are assumed.

Considering the overall energy needed to remove carbon dioxide from air (right bar chart in figure 6.3), it can be seen that lower energy estimates are able to reduce the input required to DAC plants both in term of electricity and heat, of about 30%.

Competition with Other Sequestration Options: NETs and CCS

Looking at bar chart below (Figure 6.4), the deployment of other NET technologies seem quite constant across all scenarios, except for the one that combines low energy estimate and exogenous cost reduction: indeed, in this case BECCS is highly reduced and afforestation even disappears at the end of the century, in favor of DAC1 that peaks dramatically in this decade. Generally, BECCS capture rate decreases in the last two decades of the century when applying low cost and energy assumption.

Other sequestration options, namely Carbon Capture and Sequestration (CCS) in the electricity and industrial sector, do not appear to be dramatically influenced by different cost assumptions for DAC: CCS from the electricity sector is deployed more in the mid-century in the *Low Energy* scenario, while industrial CCS results constant in all cases.
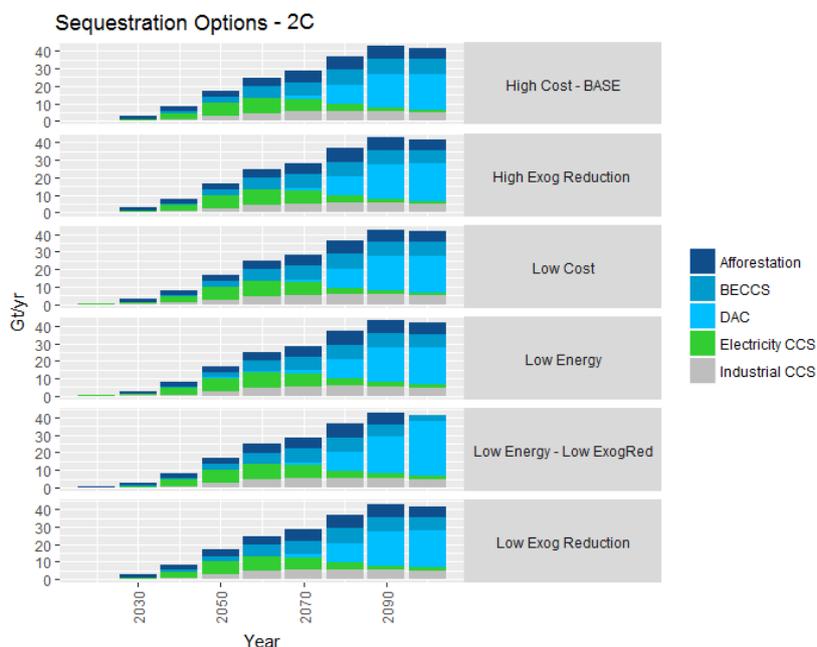
Figure 6.4: Deployment of sequestration options in 2°C, with different cost/energy assumptions.

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[3]High cost assumptions correspond to "old" estimate around 600$/ton: note that this is more a benchmark to see impact of very high cost but does not correspond to cost estimates up-to-date.

The Energy System

In order to understand the impact of different cost and energy assumptions on the energy system, the electricity mix does not appear to be very meaningful as the variability across scenarios is quite limited. Differently, more significant changes can be seen looking at the Total Primary Energy Supply (TPES), that means considering the primary energy needed to fuel the system (Figure 6.5).

It can be seen that exogenous cost reduction leads to a primary energy coming from renewable 20 EJ/yr higher than the base case in 2090-2100. On the other hand, low energy estimates increase the amount of oil (7 EJ/yr more in last decades) and coal (around 10EJ/yr more, already in mid-century from 2040) in the primary energy mix. This is evident also when reduced energy input are coupled with exogenous cost reduction, and it is associated with an increase in the role of natural gas as well (20 EJ/yr more in last decades). As before we noticed that the overall heat required as input to fuel DAC plants is reduced in these scenarios (provided in form of natural gas for DAC1 plants), this increase in the primary energy coming from fossil fuels can be related to the higher deployment reached by DAC plants. Indeed, as already discussed, capturing carbon dioxide from air allow to offset different source of emissions, therefore allowing the use of fossil resources later in the century and decreasing the need of deep decarbonization. The amount of nuclear and hydro in TPES results almost unchanged across scenarios.
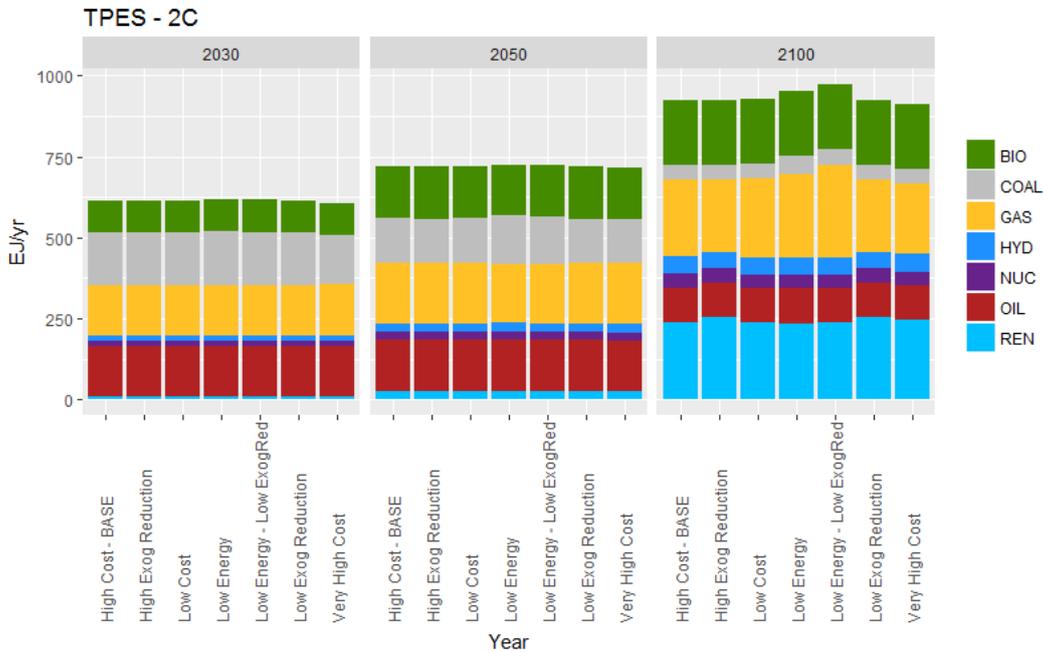
Figure 6.5: Total Primary Energy Supply [EJ/yr] for 2°C, with different cost/energy assumptions.

Even if the electricity mix does not show significant changes across scenarios, there is still an impact on the deployment of renewable capacity, mainly for solar PV and thermal (Figure 6.6). Indeed, when costs are reduced exogenously as a result of learning effects, the amount of solar thermal installed is higher from 2080 on, reflecting the increase in DAC2 capacity in the same decades. This technology requires waste heat to operate, that can be provided by solar CHP and nuclear plants: indeed also nuclear capacity results slightly higher in that period. In the same years, the amount of solar PV capacity is slightly reduced, given that solar plants can provide at the same time electricity and waste heat. Differently, when the exogenous cost reduction is combined with low energy needs, solar capacity appears to be reduced in the last decades, both for PV and for solar thermal.
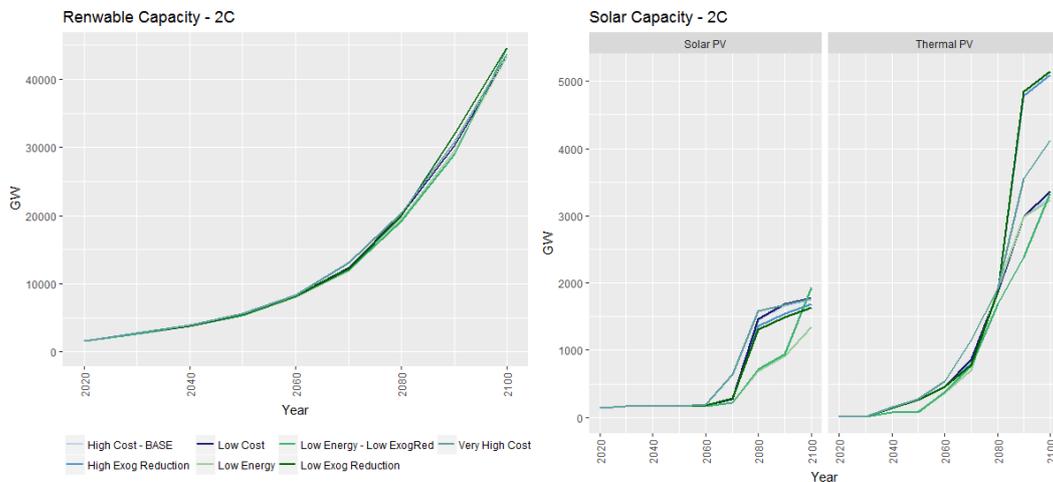
Figure 6.6: Renewable capacity installed [GW], focusing on solar, in 2° C scenarios, with different cost/energy assumptions.

Regional distribution

On a regional scale, the main impact can be seen once again in the scenario with *Low Energy* assumptions, when only DAC1 is installed worldwide. In this case, the cumulative amount of carbon dioxide captured by DAC plants raises dramatically in Asian countries (ODA, not China, where it actually decreases) and Western Europe, while also Australia, Russia and Central-South America show an marked increase.

Differently, China and Africa reveal a smaller cumulative capture, while India and Middle East completely disappear from the scene. From figure 6.7, it is interesting to notice that in these country the disappearance of DAC is not substituted by any other CDR option. For instance in India when DAC is reduced also BECCS deployment results lower, with 2-3 Gt less of cumulative capture).

On the contrary, when low energy estimates are coupled with cost reduction, the regional breakdown is more similar to the base one, given that in this scenario DAC2 starts to have a role again, balancing the ratio among the two technology options. Considering also previous results, we can conclude that there is a regional characterization of Direct Air Capture technologies, and this is especially true for DAC1.
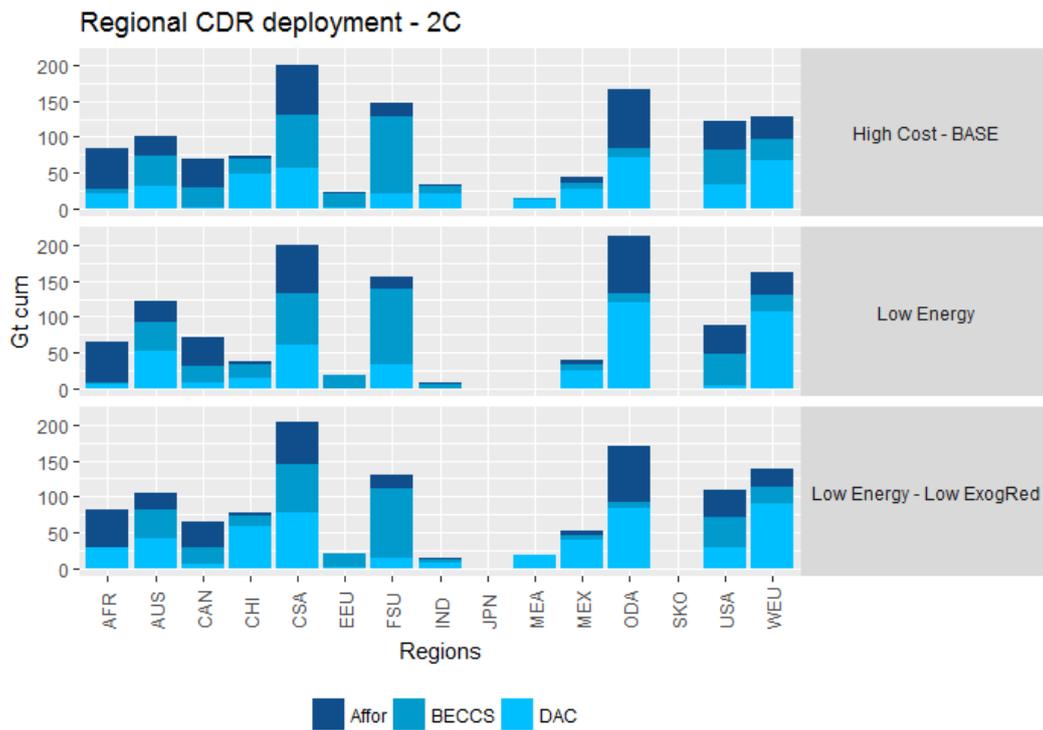
Figure 6.7: Regional breakdown of different sequestration options. Cumulative capture along the century is considered [Gt_{cum}], for 2°C scenarios, with different cost/energy assumptions.

6.1.2 Energy and Cost Assumptions in 1.5°C scenarios

The most interesting result of applying energy and cost sensitivity with a 1.5°C target, is the impact of *Low Energy* estimates: both alone and combined with exogenous cost reduction, they lead to the deployment of DAC1 exclusively (see Figure 6.8), installed mainly in Western Europe. Moreover, high cost estimates do not reduce the amount of DAC deployed in first decades of the century as it appeared before, as this technology result essential to meet such a stringent target.

While in 2°C scenarios, with Low Energy assumptions the overall energy input into DAC power plants was lower than in other scenarios, with a 1.5°C-consistent budget the reduction of specific energy inputs is not enough to counterbalance the increase in installed capacity, therefore as a net result DAC plants require more energy than before.

With a stringent target, when combining low energy parameters and exogenous cost reduction, DAC1 appears to be the most favorite option overall, while in 2°C scenarios DAC2 still holds part of the total capacity installed with the same assumptions. Moreover, capture plants based on amines do not seem to be favored in low cost scenarios as it appeared before: this can be probably explained with the fact that with such a stringent mitigation target there is no role in the electricity mix for gas-based power plants at the end of the century. Consequently, natural gas commodity may result particularly cheap as there is still lot of supply available, therefore it can be used as a inexpensive energy input for DAC plants.

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Figure 6.8: DAC deployment [Gt/yr] and energy required to operate it [EJ/yr] for 1.5°C scenarios, with different cost/energy estimates.
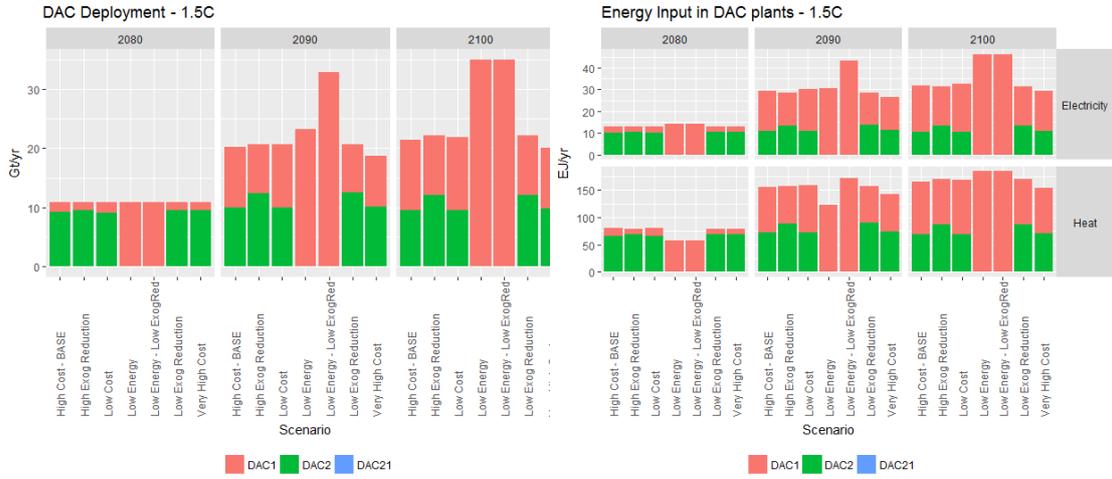
1.5°C Scenario	2070	2080	2090	2100
BASE - High Cost	1.75	10.85	20.28	21.42
High Exog Reduction	1.75	10.85	20.74	21.22
Low Cost	1.75	10.85	20.66	21.86
Low Energy - Low Exog Reduction	1.75	10.85	32.79	35.00
Low Energy	1.75	10.85	23.26	35.00
Low Exog Reduction	1.75	10.85	20.73	22.22
Very High Cost	1.75	10.85	18.71	20.10

Indeed, in these scenarios, natural gas holds a higher share of total primary energy, as it can be seen from Figure 6.9, reducing the need for biomass and renewables. It should be noted that the bioenergy potential limit is not even hit in these scenarios, that means less than 200 EJ/Year of primary energy coming from biomass.

This is reflected also in the electricity mix, as the lower share of biomass-based power plants in 2100 is counterbalanced by an increase in solar, wind and other renewable sources, while the total amount of electricity produced is almost constant across scenarios. It is interesting to notice that in 2050 coal still holds a share of the electricity production, while normally 1.5°C scenarios require a complete phase-out from coal-based power plants early in the century. Similarly, looking at the residual emissions from different sectors across scenarios, it can be seen that emission from transport, power and industrial sector are higher with *Low Energy* assumptions, as if the deployment of DAC1 technologies on a large scale (replacing BECCS) allows less urgent decarbonization from these sectors.

Considering the CO_2 captured by other CDR options in these scenarios, it can be clearly seen that DAC becomes the most convenient option to remove carbon dioxide once energy inputs are reduced, replacing also afforestation in the last decades and capturing completely the 35 Gt/yr allowed, similarly to scenarios with a 2°C target. Moreover, it should be noted that CCS in the electricity sector results slightly higher in middle century, when coal still holds a share in the electricity mix and DAC is not yet deployed.
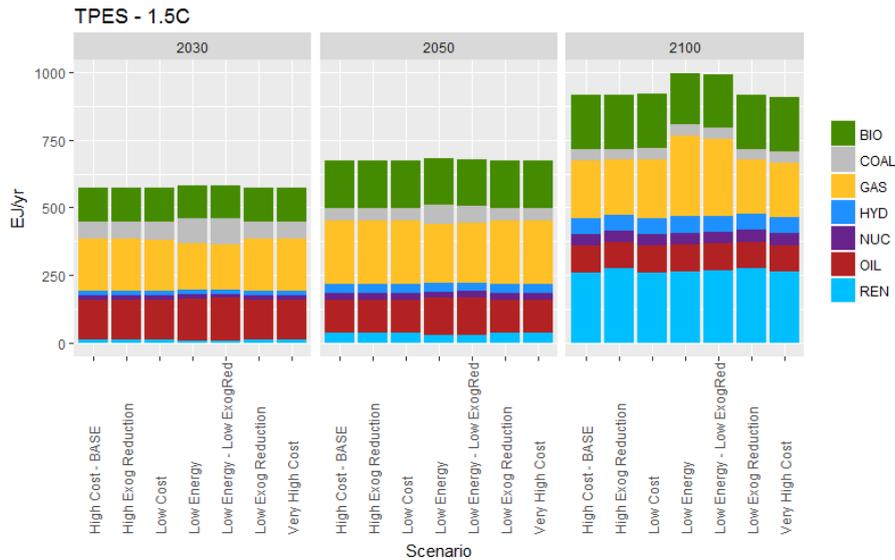
Figure 6.9: Total Primary Energy Supply [EJ/yr] for 1.5°C, with different cost/energy assumptions.

Looking at net emissions pathways (Figure 6.10), it can be seen that, even if the deployment of CDR technologies is always constrained by the 35 Gt cap applied, net emissions in scenarios with *Low Energy* assumptions results higher up to 2070, reaching more negative levels at the end of the century, as the residual emissions are reduced of about 5 Gt/yr in last decades. Emission pathway starts being different in 2080 and from 2090 the gap becomes even higher. In these scenarios, BECCS plants reduce their capture rate to almost 0 Gt/yr: this suggest a sort of competition or incompatibility between DAC1 and BECCS. Moreover, the carbon price results about 1000 $/ton lower.

In conclusion, it seems clear that the main obstacle for diffusion of DAC1 is the energy energy requirement, while for DAC2 is related to cost estimates. Indeed, there is higher potential for cost reduction related to this technology as design of plants can be developed and optimized for this application. Differently, for DAC1 the majority of equipment needed comes from well-known processes, as the pulp and paper industry, so that it may decrease cost more rapidly but with a limited reduction in time, that means a higher floor cost eventually reached.

Net emissions - 1.5C
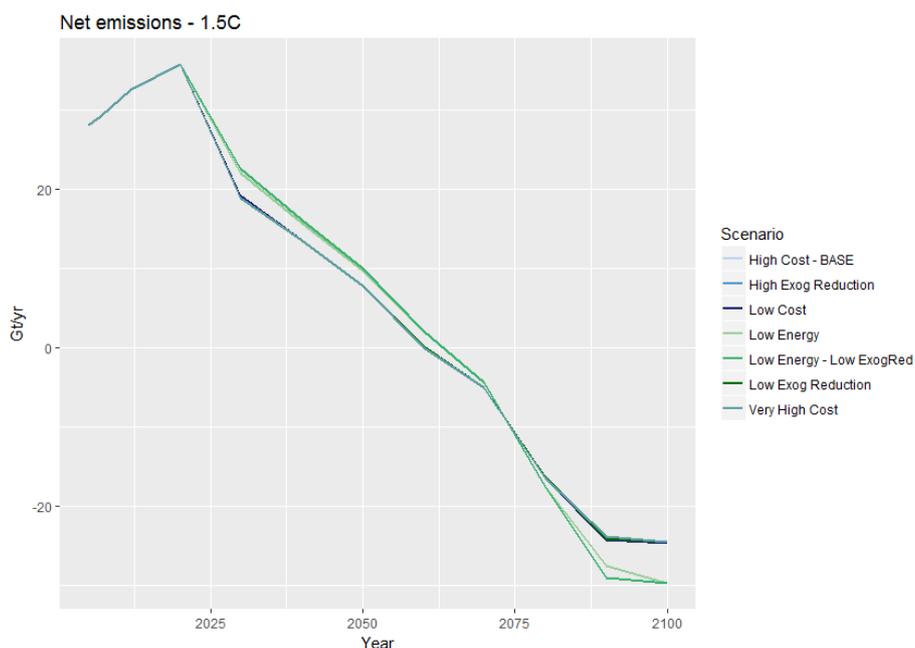
1.5°C Scenario	2020	2030	2040	2050	2060	2070	2080	2090	2100	%
BASE - High Cost	24	143	233	380	619	1008	1642	2675	4357	
High Exog Reduction	24	143	233	379	618	1006	1639	2669	4348	-0.2%
Low Cost	24	143	233	380	619	1098	1644	2677	4361	+0.1%
Low Energy	24	111	181	294	479	780	1271	2070	3372	-23%
Low Energy - Low Exog Reduction	24	106	173	282	459	747	1217	1983	3230	-26%
Low Exog Reduction	24	143	233	379	618	1006	1639	2669	4348	-0.2%
Very High Cost	24	144	235	382	622	1014	1651	2690	4382	+0.6%

Figure 6.10: Net emission and carbon price for 1.5°C, with different cost/energy assumptions.

6.1.3 Further Analysis

Removing the Growth Constraint

As we discussed previously, the impact of energy and cost is quite limited due to the binding growth constraint applied. Therefore, I removed it temporarily for the 2°C target, to understand how the model would behave in response of different technical parameters for DAC.

As expected, removing the growth constraint, more marked differences can be seen across scenarios, especially in the timing of DAC deployment: indeed, without imposing a limit on the annual growth, cost assumptions are the main parameter influencing the starting year for the deployment of this technology, with around 8 Gt/yr capture already in 2060 when costs reduce throughout the century thanks to learning effects. Differently, the final extent reached does not change much across scenarios, with absolute differences lower than 1 Gt/yr. Earlier in the century, the capture capacity installed is based almost exclusively on amine-modified adsorbent (DAC2) and its capture rate remains almost constant along the century, while DAC1 is being deployed only after 2080 (figure 6.11). It can be still found that low energy assumption favor greatly DAC1, while reducing the cost result in a higher capacity of DAC2 plants.
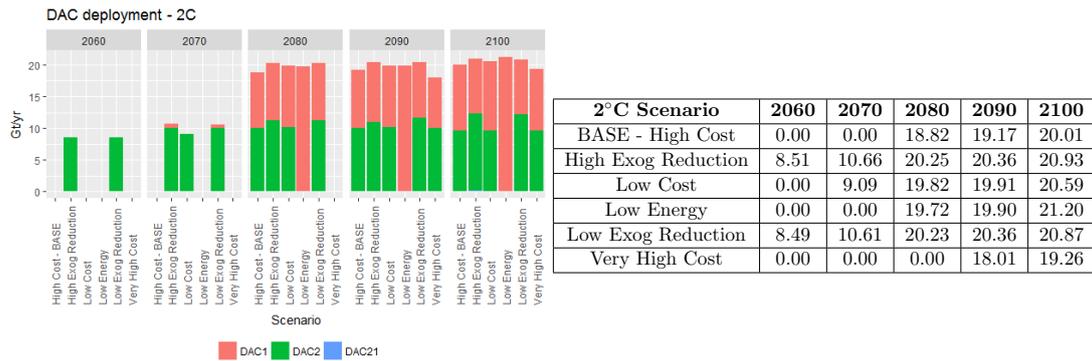
Figure 6.11: DAC deployment [Gt/yr] and energy required to operate it [EJ/yr] when no growth rate is applied for 2°C scenarios, with different cost/energy estimates.

Looking at the carbon price, it is interesting to notice that they are generally smaller when no growth constraint is applied: in particular, while previously *LowEnergy* assumptions led to the smallest marginal abatement cost (-15% with respect to base case), here the lowest carbon price appears in both scenarios with an exogenous cost reduction, with around 27% reduction with respect to the base case. Indeed, these are the scenarios allowing a more smooth growth of DAC throughout the century, with a resulting annual growth rate around 30%, still feasible when compared to historical technology diffusion paths.

Generally, the trends are similar to what has been discussed before, but the differences are by far less marked when the growth constraint is applied, as the deployment of DAC plants is not allow to differ much across scenarios, therefore reducing the impact on the overall system.

Different Cost Reduction Rates

As discussed previously in Section 4.1.4, technical learning can lead to different rates of cost reduction, therefore an additional sensitivity has been applied to this parameter, considering a range between 1 and 15% annual reduction.

The overall impact appears almost negligible when a growth constraint is applied, both with 2°C and 1.5°C target, with a small difference only on the share of different DAC options. In particular a lower cost reduction rate (1% per year) increases the capture rate from DAC1 of about 2-3 Gt/yr at the expense of DAC2, in line with DAC2 deployment being more sensitive to cost reduction, as discussed before. As a consequence in the last decades the capacity of solar thermal installed is reduced, as well as nuclear, being the technologies that provide waste heat for DAC2.

It should be noted that with 6 and 15% exogenous reduction rate, results are almost the same, meaning that the model does not care whether the floor cost is reached already in 2030-2040 (15%) or slightly later around 2050-2060 (6%), given that DAC deployment takes place at a Gt scale only from 2070 on

Role of Waste Heat to Fuel DAC2

Generally, DAC21 is not deployed by the model when a full portfolio of NET options is available, as it is not convenient to produce heat on purpose to be fed to amine-based DAC plant: considering also that a cumulative cap is applied, the model is forced to allocate this capacity only to the most competitive alternatives. Therefore, it would be interesting to understand what happens if the capture technology employing amine adsorbents can not be fuelled by cheap waste heat any longer. This case has been explored across different energy and cost scenarios, removing the technology DAC2.

Both in 2°C and 1.5°C the capacity of DAC2 is now taken by DAC1 that becomes the only air capture technology deployed by the model, underlining that it is not convenient at all to produce that amount of heat on purpose. Indeed, in TIAM model the heat commodity used as input to amine-based plants comes mainly from biomass-fired CHP plants: as a consequence, DAC21 results to be in competition with BECCS for the use of bioenergy, with a lower efficiency of the entire supply chain to capture the same amount of CO_2.

Looking at carbon price, the difference is negligible, about 1%, and also the net emission pathways are not highly influenced. Considering the entire energy system, there is a significant impact on the renewable capacity installed, being about 10% more in 1.5°C scenarios and only 4% in 2°C ones: this is mainly evident for solar, as without DAC2 almost 1000 GW more are installed in the 1.5°C scenarios (in 2°C it is about 300 GW more). Differently, other sequestration options do not results to be affected by this "internal" change among DAC technologies

6.2 Sensitivity on the Time Discount Rate

Now, it will be explored the impact that the discount rate applied by the model to future costs may have on the development of DAC and the mitigation pathway undertaken, both when DAC is the only NET option available and when it is part of a wider CDR portfolio, with a 2°C and a 1.5°C target.

This parameter captures the willingness to transfer mitigation costs to future generations that will face higher climate damages: if the discount rate, also referred to as social rate of time preference, is set to zero, it means that future welfare and future costs will be equally important as the current ones, therefore emission reduction is likely to be more evenly distributed across the time horizon. Differently, with a higher discounting, future costs are less relevant in the overall objective function, therefore mitigation is likely to be deferred to the last decades of the century.

Note that in TIAM there are two different discount rates defined within the model: the *general discount rate*, that represent the discounting to the base year, and a *technology-specific discount rate*, representing how the payment of any capital cost is spread over the economic life of a plant, so to be annualized. I am considering the former one, changing it from the default value of 5% to a higher one (10%) and to a value close to 0% (0.01% precisely) so to have almost no discounting of future time periods. Energy and cost assumptions are the ones of the baseline scenario, being high constant estimates.

Scenarios Investigated:

|	Discount Rate		Full NET portfolio	Only DAC
	Low	0%	✓	✓
2°C	Baseline	5%	✓	✓
	High	10%	✓	✓
	Low	0%	✓	✓
1.5°C	Baseline	5%	✓	✓
	High	10%	✓	✓

6.2.1 Impact of Discount Rate in 2°C Scenarios

Generally, the lower the discount rate, the sooner mitigation takes place, so to avoid more drastic (therefore expensive) emission reduction in the last decades.

Looking at the net emission pathways in Figure 6.12, this can be clearly seen in scenarios with a full NET portfolio, where with a 10% discount rate emissions peak later, in 2030, and then decline rapidly around 2080. Differently, without any discount of future time periods, more drastic decarbonization takes place between 2020-2030, with an annual emission reduction rate around 9%, then flattening to less than 0.5 Gt reduction per year. This leads to a level of net emissions around -7 Gt/yr at the end of the century, being much higher than all other scenarios with DAC availability, where negative emissions would lead up to -22 Gt/yr .

Considering scenarios where only DAC is available as a CDR option, the difference when changing the discount rate is significant only in the first half of the century, as from 2060 the emission pathways does not change much. This suggests that the model is treating DAC as a backstop technology, given that it is the only alternative present, thus deploying it as much as possible when it becomes economically available: with high constant costs and high energy inputs, it represents a convenient mitigation option only in the second half of the century, regardless of the emission pathways followed up to that moment. Indeed, in 2030 and 2040 the scenario with lower discount rate shows residual emission level much lower than the 5% and 10% discounted scenarios, with about 6 and 13 Gt/yr less respectively. After 2050 this gap shrinks to less than 2 Gt/yr. Differently, the availability of a diversified mitigation portfolio including also BECCS and afforestation, allows to spread the mitigation burden more evenly across the century according to the discounting applied to future time periods.

The difference in the net emission level across 'only DAC' scenarios up to 2050 still has an impact on the values of marginal abatement cost (i.e. carbon price), as expected. Indeed, in all scenarios, reducing the discount rate to almost zero value makes the carbon price about 70/80% lower, while increasing it to 10% leads to a carbon price up to 10 times higher by the end of the century (see table 6.12).

Figure 6.12: Net emission pathways and carbon price for 2° C, with different discount rates.
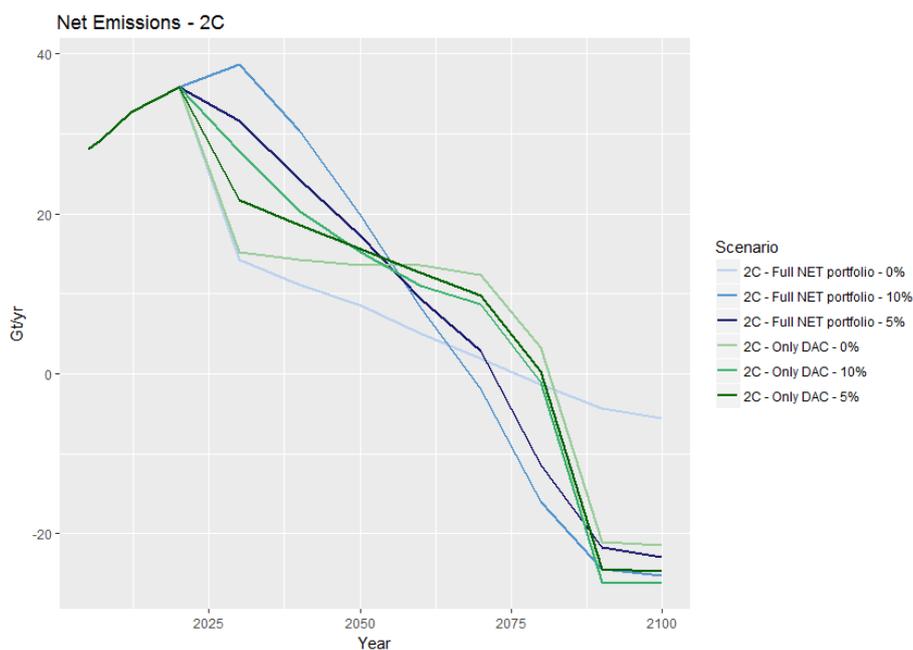
2°Scenario	2020	2030	2040	2050	2060	2070	2080	2090	2100
Full NET Portfolio - 0%	11	322	323	323	323	324	324	324	325
Full NET Portfolio - 10%	48	13	34	89	231	599	1552	4027	10444
Full NET Portfolio (5%)	24	41	66	107	175	285	465	757	1233
Only DAC - 0%	11	606	607	608	608	609	609	610	611
Only DAC - 10%	48	81	210	544	1410	3657	9485	24603	63814
Only DAC (5%)	24	168	274	446	727	1184	1929	3141	5117

Similarly, the total system cost is reduced when future time periods are discounted less, ending up with a mitigation cost of only 1% of global GDP. This is true when a full portfolio of NET is available, while with only DAC, mitigation cost still accounts for 3-4% of global GDP as a massive deployment of DAC takes place in the last decades. With a higher discount rate, mitigation costs in last decades can reach 5% of projected global GDP, up to 10% when only DAC is available.

DAC Deployment and Other Sequestration Options

The most visible impact of the discount rate on DAC deployment can be seen when a full portfolio of NET is available for the model (see figure 6.13a): indeed, with a lower discounting, only less than 8 Gt/yr of CO_2 are captured by DAC at the end of the century, compared to about 20 Gt/yr in the base scenario (5% discount rate). As noticed before, the lower the discounting applied to future time periods, the greater the mitigation efforts in the first decades to decarbonize the energy system, leading to a reduced need for negative emissions technologies. Globally, NETs are able to capture only around 22 Gt/yr by the end of the century, summing over DAC, BECCS and afforestation capacity, not even hitting the constraint. In this scenario, only DAC2

is installed, while with the highest discount rate DAC1 results again favored against DAC2, as its higher costs are greatly discounted in future time periods within the model. Note that the difference between 5 and 10% case is almost negligible in term of overall capture capacity, but the share of different DAC options results affected.

As already highlighted before, the impact of the discount rate is different when no BECCS or afforestation can be deployed: limiting the number of alternatives to reduce carbon concentration in the atmosphere, the model still relies a lot on the capture rate provided by DAC plants, which are installed with similar capacity regardless of the discount rate. Even if the overall deployment does not change, still an impact can be seen in term of share of technologies, with high discount rates favoring DAC1.

2°C Scenario	2060	2070	2080	2090	2100
Full NET - 0%	0.28	1.75	3.35	5.98	7.65
Full NET - 10%	0.28	1.75	10.85	20.32	21.84
Full NET (5%)	0.28	1.75	10.85	19.15	20.04
Only DAC - 0%	0.28	1.75	10.85	35.00	35.00
Only DAC - 10%	0.28	1.75	10.85	35.00	35.00
Only DAC (5%)	0.28	1.75	10.85	35.00	35.00

(a) DAC Deployment

(b) Other Sequestration Options
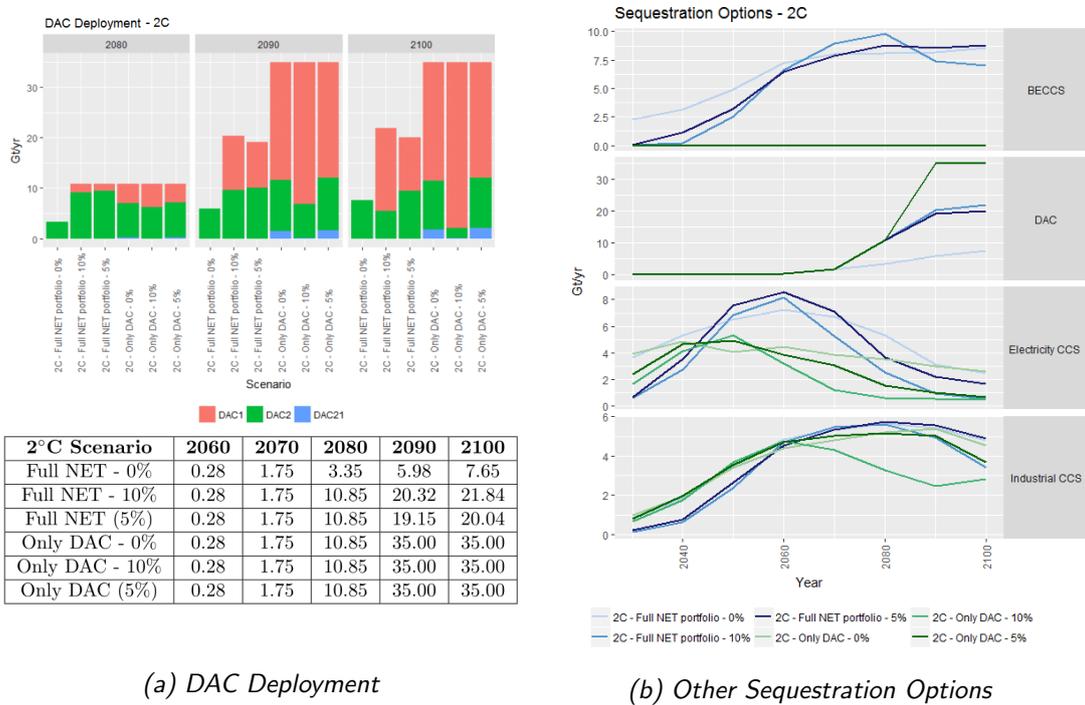
Figure 6.13: DAC deployment and other sequestration options for 2°C, with different discount rates.

The role of BECCS (Figure 6.13b, in scenarios where it is available) is higher in the first decades when we apply a smaller discount rate, as DAC technologies are not enough mature and economically convenient to be deployed in the first half of the century. As BECCS capture rate remains around 8 Gt/yr from 2070 on, NET capacity results almost equally divided among the three options (bio-energy, air capture and afforestation), without a marked prevalence for DAC as it appears with a 5% discount rate. This suggests that DAC technologies will be needed more if mitigation is being delayed in time, while more drastic short-term efforts could reduce the need for massive deployment of this technology and corresponding higher mitigation cost. Nevertheless, even when drastic emission reduction is applied in the first decades, DAC will still play a role in the second half of the century to reduce the impact of mitigation on the energy system, requiring less intermittent renewable generation.

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Looking at other sequestration options (Figure 6.13b), it can be seen that CCS from industrial sector does not result much influenced by the discount rate, being slightly reduced in the last decades when a higher discounting is applied. Differently, in the electricity sector it holds more stable role throughout the century when future is discounted less, while with higher discounting its capture capacity peaks it 2050-2060 and then declines rapidly as most of the mitigation effort is taken by (expensive) direct air capture plants.

It is interesting to notice that the smaller the discount rate, the greater the role of coal-based CCS power plants, even later in the century, and the smaller the capacity of gas-based ones with respect to base 5% discounted scenarios. On the other hand, higher discount rates lead to almost no CCS from coal plants in the second half of the century and still to a reduced role for gas-based power plants due to the large gas needed by DAC1 plants. This is true both with a full NET portfolio and when when only DAC is available.

The Energy System

The extent to which future time period are discounted, and the consequent deployment of DAC and other CDR technologies, do have an impact on how the energy system will look like across the century, in particular considering the renewable capacity installed (see Figure 6.14). Indeed, the higher the discount rate, the larger the capacity of solar, hydro and other renewable that needs to be installed from 2050 on, in order to decarbonize rapidly the system and meet the climate target. Differently, lower discount rates applied to future decades lead to higher deployment of mitigation options (both electricity CCS and CDR) earlier in time, thus reducing the role of renewable capacity, and therefore also the share of intermittent generation in the electricity system.
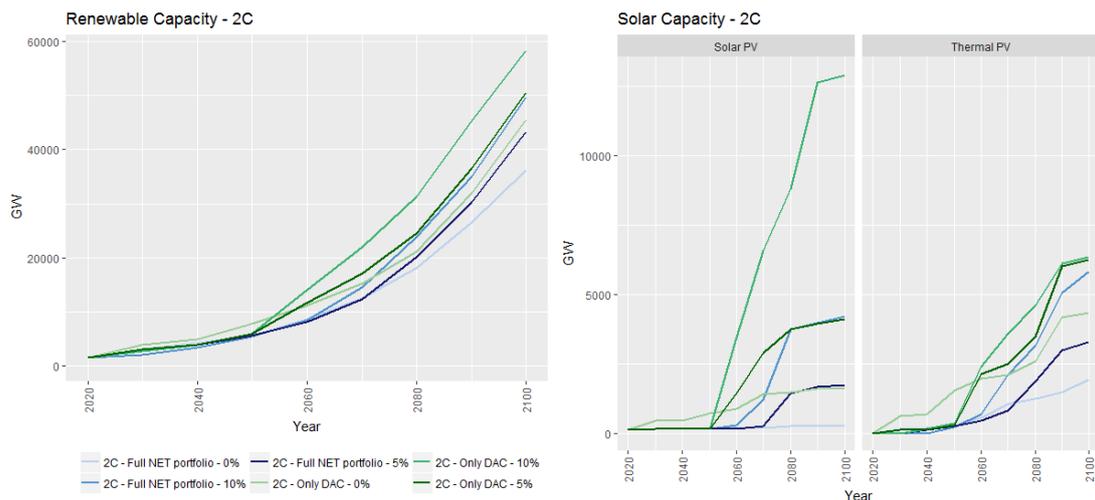
Figure 6.14: Renewable capacity installed [GW], focusing on solar, in 2°C scenarios, with different discount rates.

Looking at the electricity production (Figure 6.15a), it can be seen that the lower the discounted rate the greater the role of gas in the electricity mix already in 2050, with a smaller share of coal. Moreover, the overall electricity production results higher in the mid-term (i.e. 2050), given that electrification is one of the strategies to decarbonize the system, and lower in the long run with respect to scenarios with higher discount rates. The share of intermittent renewable generation is reduced of about 10 percentage points going from high to low discount rates.

(a) Electricity Mix [EJ/yr]   (b) Sector Emissions [Gt/yr]
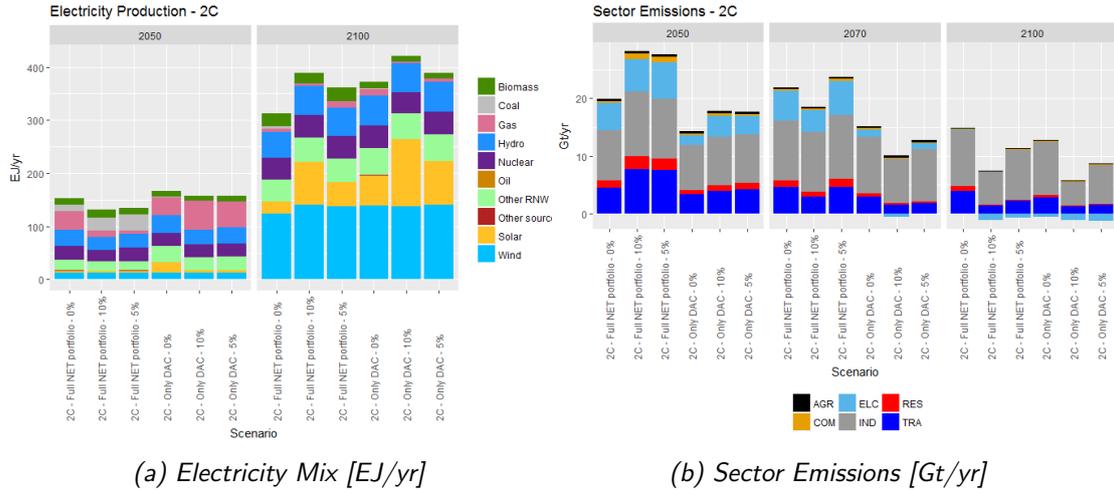
Figure 6.15: Electricity mix and sector emissions for $2°C$, with different discount rates

The chart in Figure 6.15b confirms what has been described before: the level of emission from different sectors is much reduced in 2050 with a lower discount rate and no drastic reduction are needed in the last decades. Looking at the origin of these residual emissions, it can be seen that with smaller discount rate (therefore lower amount of DAC installed), transport, power and industrial sectors undergo more drastic decarbonization around 2050, then remaining almost stable across the following decades. Differently, with higher discounting, emissions from the industry and the energy system, representing the most carbon-intensive sectors, are strongly reduced between 2070 and 2100.

Regional Distribution

While the regional amount of net emission does not change dramatically across scenarios, some regions appear to switch their removal strategy when different discount rates are applied: indeed, when moving from 5 to 10% discounting, Western Europe (WEU) deploys less CCS in electricity sector and more DAC capacity, while China (CHI) reduces BECCS plants to favor more DAC and at same time emits more CO_2. When only DAC is available, it is interesting to notice that it is being installed mainly in Former Soviet Union region (FSU), Western Europe and China as soon as the discount rate is increased. This reflects the common pattern of regional distribution already highlighted in previous sections.

6.2.2 Impact of Discount Rate in 1.5°C Scenarios

With a more stringent mitigation target, the discount rate does not have a significant impact on the net emission pathways when only DAC is available to remove carbon dioxide from the atmosphere (Figure 6.16), while the resulting carbon price for highly discounted case result extremely high, thus showing infeasibility for this scenario. On the other hand, the effect of different discounting is more marked when the full portfolio of NET alternatives can be deployed. Differently from the 2°C results, here the net level of emissions result highly negative across all scenarios, with more drastic reduction reached when no BECCS or afforestation are available, up to -25 Gt/yr.
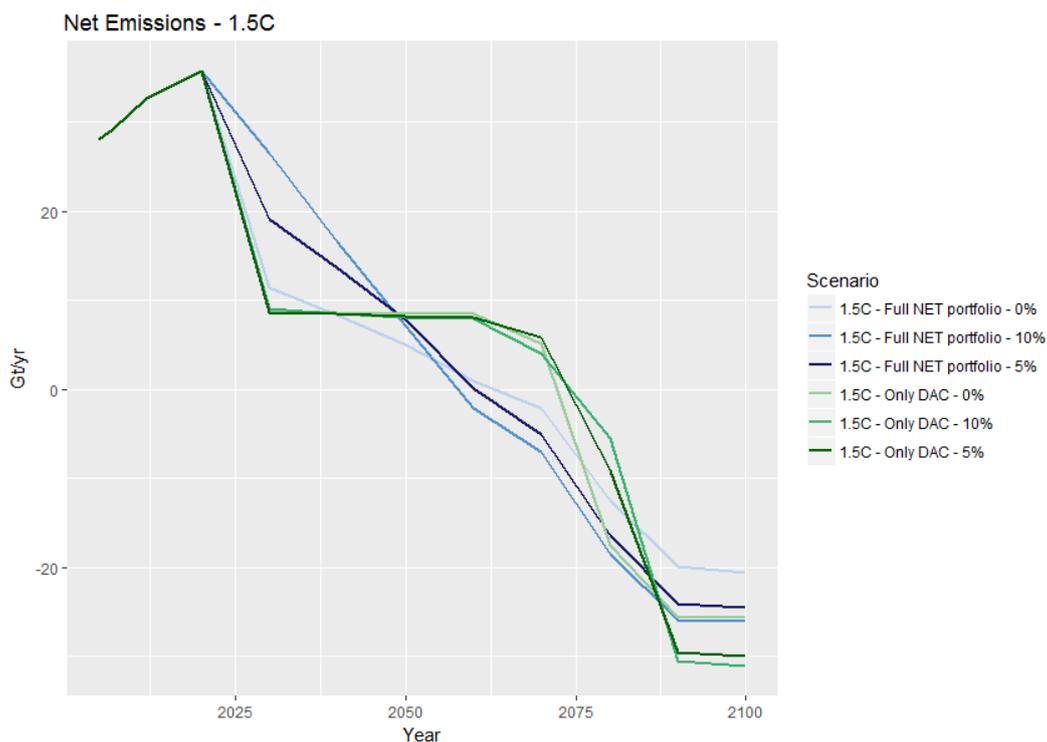
1.5°C Scenario	2020	2030	2040	2050	2060	2070	2080	2090	2100
Full NET Portfolio - 0%	11	528	529	529	530	530	531	531	532
Full NET Portfolio - 10%	48	68	176	455	1181	3062	7943	20602	53437
Full NET Portfolio (5%)	24	143	233	380	619	1008	1642	2675	4357
Only DAC - 0%	11	9179	9188	9197	9206	9216	9225	9234	9243
Only DAC - 10%	48	4630	12009	31149	80794	209558	543539	1409800	3656658
Only DAC (5%)	24	8262	13458	21921	35708	58164	94743	154326	251381

Figure 6.16: Net emission pathways and carbon price for 1.5°C, with different discount rates.

DAC Deployment and Other Sequestration Options

Looking at the deployment of DAC with different future discounting in Figure 6.17a, it can be noticed that it is affected earlier in the century when this is the only CDR option, with capture rate being equal to 27 Gt/yr already in 2080 when a 0% discount rate is applied. It is interesting to notice that in this case DAC21 plays a role to meet

such a stringent target, while it is completely absent in other scenarios. Differently from the 2°C results, here a lower discount rate is not able to reduce the amount of negative emission needed to meet the mitigation target.

Once again, a higher discount rate favor the deployment of DAC1 over DAC2, even more than with a 2°target: indeed, with 10% discounting, only plants based on hydroxide solutions are deployed in 2100, and across all scenarios DAC1 is present while DAC2 appears to be often overtaken by the other capture technology.

1.5°C Scenario	2060	2070	2080	2090	2100
Full NET - 0%	0.28	1.75	10.85	18.66	19.77
Full NET - 10%	0.28	1.75	10.85	20.85	23.60
Full NET (5%)	0.28	1.75	10.85	20.28	21.42
Only DAC - 0%	0.71	4.39	27.20	35.00	35.00
Only DAC - 10%	0.28	1.75	10.85	35.00	35.00
Only DAC (5%)	0.46	2.87	17.74	35.00	35.00
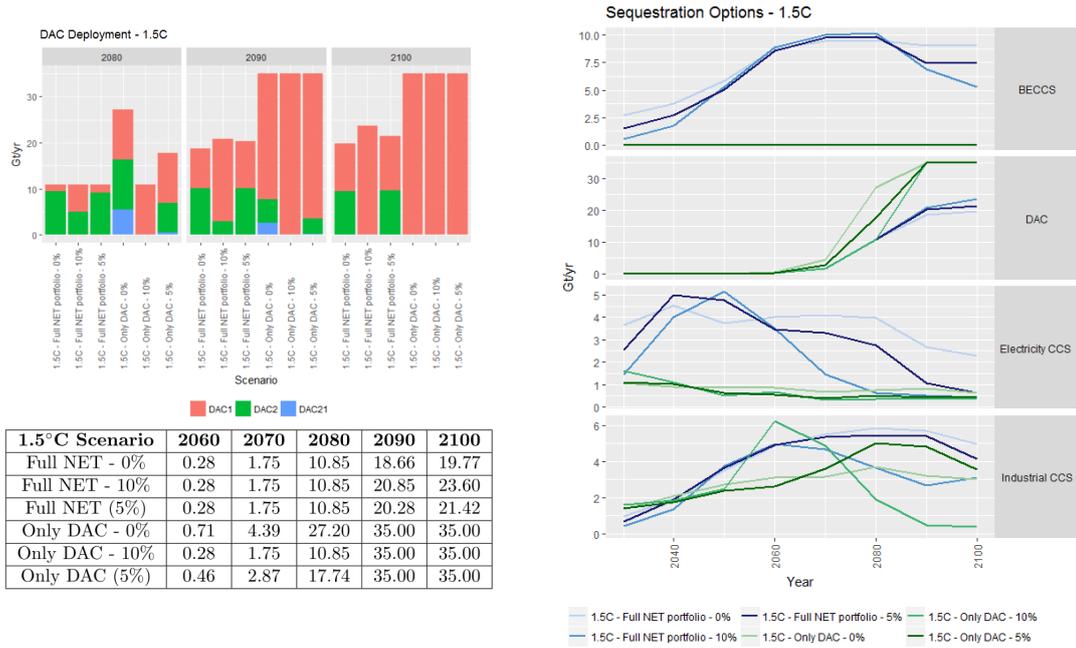
(a) DAC Deployment

(b) Other Sequestration Options

Figure 6.17: DAC deployment and sequestration options for 1.5°C, with different discount rates

Looking at Figure 6.17b, it can be noted that when DAC is the only removal option, sequestration is being reduced also in other sectors with respect to scenarios with a full selection of NETs, as the level of residual emission is generally lower and the energy system is being highly decarbonized to meet the target, already in 2050 (compare with Figure 6.18b). This is significant mainly for CCS in the electricity sector, regardless of the discounting applied. BECCS deployment appears to be much less influenced by different discount rate than in 2°C scenarios, suggesting that with a more stringent target the alternatives to achieve a net removal of carbon dioxide from the atmosphere needs to be deployed at their maximum potential. Similarly to what has been noticed previously, the deployment of CCS is more constant along the century with lower rate, while it peaks around mid-century when future is discounted more.

The Energy System

Looking at the charts in Figure 6.18 representing the energy sector, it can be seen once again that a lower discount rate requires a more drastic decarbonization of the

power sector in the short term: while with a 2°C target, this was reflected by a larger share of gas with respect to coal, with 1.5°C renewables cover a larger share in the electricity mix already in 2050. Once again, in the long run a massive deployment of renewable capacity is required in scenarios with higher discounting, in order to counterbalance the delayed mitigation efforts during the first half of the century (Figure 6.18c): solar PV is the low-carbon technology which is installed the most from 2050. Generally the amount of electricity produced is higher than in 2°C scenarios.

Considering sector emissions (Figure 6.18b), the trends are similar to the one highlighted for 2°C results, with more drastic emission reductions required in the second half of the century when larger discount rates are applied. As discussed before, with a 1.5°C target and only DAC as a CDR technology, the impact of the rate of time preference is limited on the level of residual emissions, with negative emissions achieved in the energy sector across all scenarios already in 2050, but by the end of the century the effect is much larger than for the 2°C case.

(a) Electricity Mix [EJ/yr]

(b) Sector Emissions [Gt/yr]

(c) Renewable capacity installed [GW]
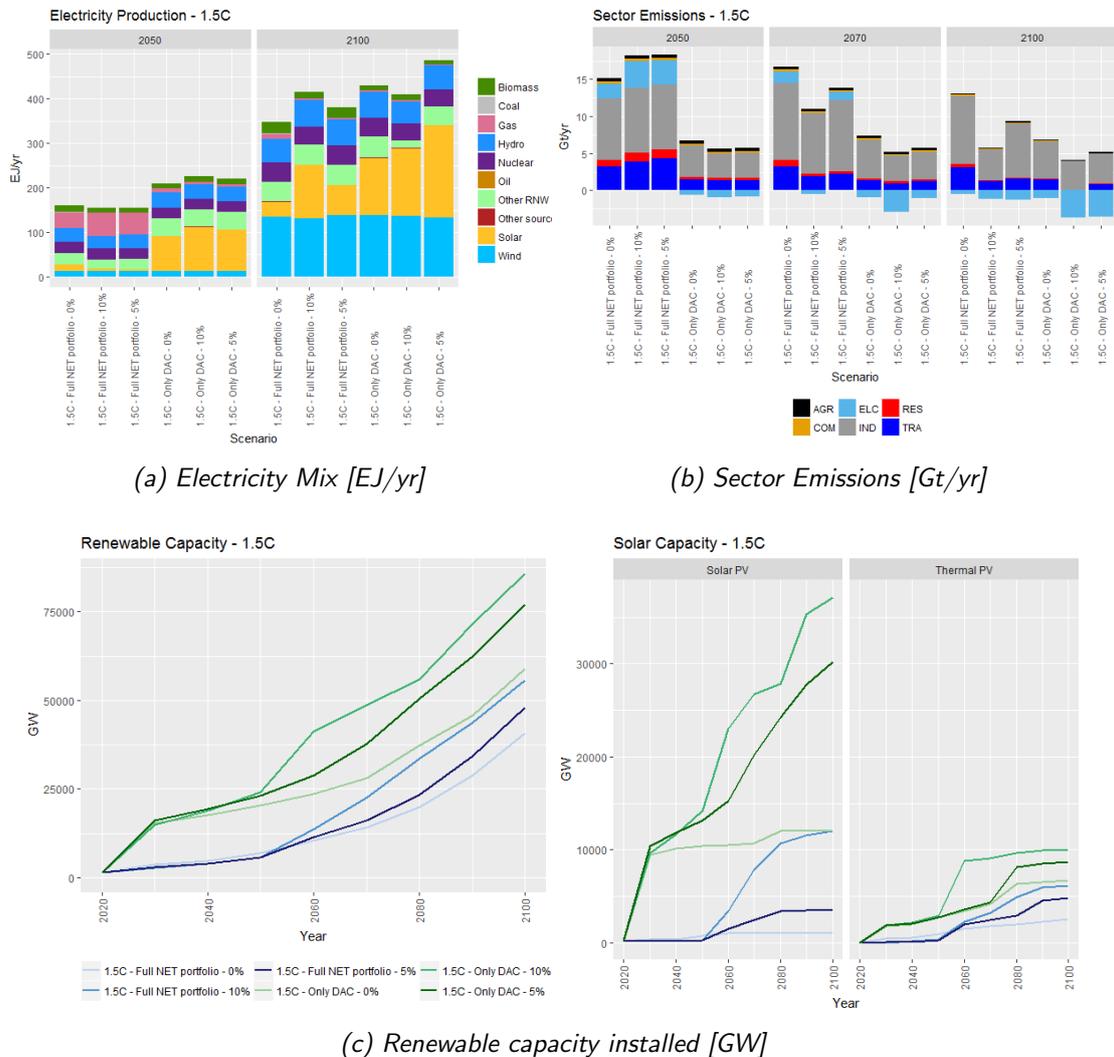
Figure 6.18: Energy production and sector emissions for 1.5°C, with different discount rates.

To conclude, all results seem to suggest that DAC1 is the option chosen by the model when mitigation is being delayed (i.e. highly discounting the costs for future generations), while if future time periods are less discounted a diversified mix of DAC technologies would be considered.

6.3 Sensitivity on Growth Constraint

As we have highlighted frequently, growth constraint results to be binding in determining the model results, therefore sensitivity analysis is needed on these parameters, to understand to which extent they are influencing the deployment of DAC technologies forecasted by the model. Both the annual growth rate [%/year] and the maximum capture capacity allowed [Gt/yr] are changed to check the model robustness.

6.3.1 Annual Growth Rate

Different growth rates have been applied, starting from a 10-15% as the lower cases, going up to 30%, according to historical comparison discussed previously in section 4.2. Since a 10% annual growth brings to levels of DAC capacity at the end of the century too small to be a relevant mitigation strategy (around 1 Gt/yr), only scenarios with 15-20-30% will be discussed, considering both base cost assumptions and exogenous cost reduction, for 2°C and 1.5°C targets.

Scenarios Investigated:

	Annual Growth		BASE - constant cost	Exog Cost Reduction
	Low	10%	-	-
		15%	✓	✓
2°C	Baseline	20%	✓	✓
	High	30%	✓	✓
	Low	10%	-	-
		15%	✓	✓
1.5°C	Baseline	20%	✓	✓
	High	30%	✓	✓

Net Emission Pathways and Carbon Price

The first thing to be noted is that the growth constraint does not affect much the extent to which DAC is deployed in 2100, being always around 20 Gt/yr out of 35 Gt/yr of CDR, but the impact is more evident on the deployment during previous decades, especially around 2070-2080. In order to counterbalance the lower capture capacity by Direct Air Capture in these decades, net emissions result lower when a 15% rate is applied, while higher rates allow to delay mitigation after 2070: this applies to both 2°C and 1.5°C target, and it is much more evident with an exogenous cost reduction and with a more stringent budget (see Figure 6.19).

(a) 2°C scenarios   (b) 1.5°C scenarios
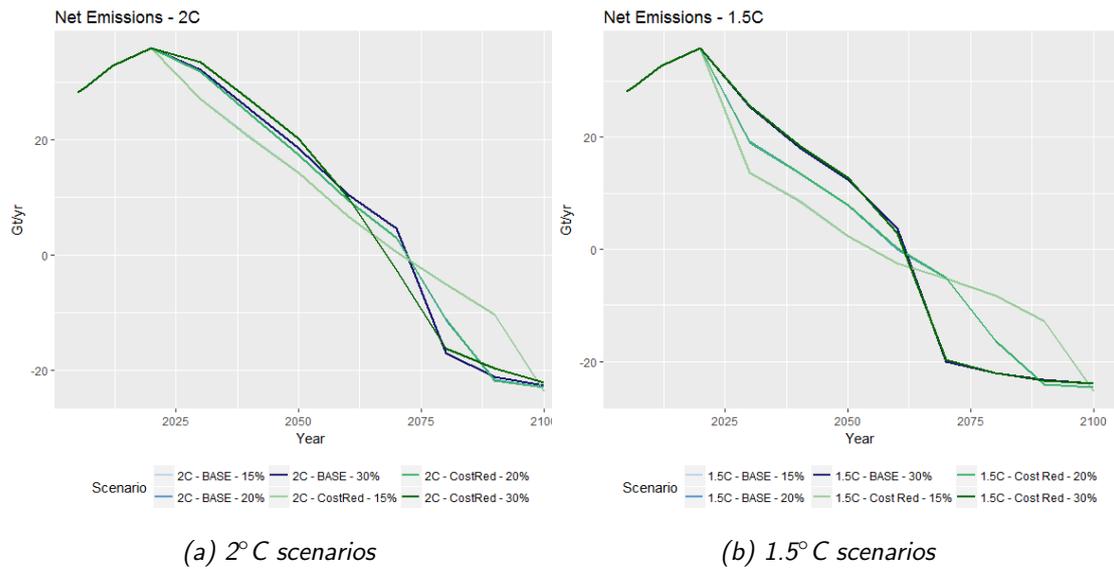
2°C Scenario	2020	2030	2040	2050	2060	2070	2080	2090	2100	%
BASE - 15%	24	66	108	176	286	466	759	1237	2014	+63%
BASE - 20%	24	41	66	107	175	285	465	757	1233	
BASE - 30%	24	36	59	97	158	257	418	682	1110	- 10%
Cost Red - 15%	24	66	107	175	285	464	756	1231	2006	+63%
Cost Red - 20%	24	40	66	107	174	284	462	753	1227	
Cost Red - 30%	24	28	46	75	122	199	324	528	860	-30%

1.5°C Scenario	2020	2030	2040	2050	2060	2070	2080	2090	2100	%
BASE - 15%	24	327	533	868	1414	2303	3752	6112	9955	+128%
BASE - 20%	24	143	233	380	619	1008	1642	2675	4357	
BASE - 30%	24	79	129	210	342	558	908	1479	2410	-45%
Cost Red - 15%	24	329	535	872	1420	2313	3768	6138	9998	+130%
Cost Red - 20%	24	143	233	379	618	1006	1639	2669	4348	
Cost Red - 30%	24	77	125	204	333	542	883	1438	2342	-46%

Figure 6.19: Net emission pathways and carbon price for 2°C and 1.5°C, with different growth rates

Considering the marginal abatement cost, we can notice that a less rapid deployment of Direct Air Capture plants leads to an increase in the carbon price around 60% and 130%, in 2°C and 1.5°C scenarios respectively. On the contrary, the impact of higher diffusion rates is not symmetric, but is more limited, leading to a reduction between 10 and 45%. It should be noted that with a 2°C target the impact of a high growth rate is more significant when the cost reduction is applied, while in 1.5°C scenarios we cannot see any difference: this can be related to the larger deployment of (more expensive) DAC1 plants over DAC2 ones.

DAC Deployment and Other Sequestration Options

As expected, the higher the annual growth rate allowed, the greater the capacity that can be installed in earlier decades: indeed, the growth constraint result binding only in the first years of DAC adoption, not at the end of the century. Few differences can be noted between 2 and 1.5 °C results, except that a more stringent target, when a 30% growth rate is allowed, requires a huge installation of DAC plants earlier in the century, reaching around 20 Gt/yr capture rate already in 2070. Differently, in 2°C scenario with exogenous cost reduction only 10 Gt/yr are captured in the same decade, provided mainly through amine-based plants.

It is interesting to notice that a higher growth rate does not imply the deployment to be shifted later in time with a more drastic ramp-up, but an early diffusion reaching large high capacity as soon as the growth constraint does allow it. It should be noted that the 35 Gt cap for CDR removal is always hit regardless of the growth constraint applied[4]: with 15% growth rate on DAC this is true only in the very last decade, while with higher growth rate it is reached already in 2070.

One interesting aspect of growth rate sensitivity is in the share between DAC technologies: generally, the higher the growth rate, the more DAC2 is installed earlier in the century, being favored over DAC1, while with smaller growth rate brings more DAC1 to be installed. This appears clearly looking at 1.5°C results, where the growth constraint is a more influencing factor, while with a less stringent target cost parameters are more relevant so that a cost reduction still favors DAC2 in the last decades also when a smaller growth rate is applied (Figure 6.20.

2°C Scenario	2060	2070	2080	2090	2100
BASE - 15%	0.09	0.36	1.44	5.82	20.43
BASE - 20%	0.28	1.75	10.85	19.15	20.04
BASE - 30%	0.09	1.26	17.43	19.20	19.99
Cost Red - 15%	0.09	0.36	1.44	5.82	20.98
Cost Red - 20%	0.28	1.75	10.85	19.70	21.21
Cost Red - 30%	2.82	10.73	20.13	20.27	20.92

1.5°C Scenario	2060	2070	2080	2090	2100
BASE - 15%	0.09	0.36	1.44	5.82	21.6
BASE - 20%	0.28	1.75	10.85	20.28	21.42
BASE - 30%	1.63	20.04	18.80	19.43	20.84
Cost Red - 15%	0.09	0.36	1.44	5.82	22.11
Cost Red - 20%	0.28	1.75	10.85	20.74	22.22
Cost Red - 30%	2.82	20.82	19.96	20.82	22.17

Figure 6.20: DAC deployment [Gt/yr] in 2°C and 1.5°C scenarios, with different growth rates.

Considering other sequestration options (Figure 6.21), it should be noted that in the

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[4]This was not true when the smallest growth rate of 10% was applied.

2°C case CCS in the electricity sector shows a positive correlation with DAC capacity, as it is deployed more in scenarios where more direct capture plants are installed. This relation can be explained by the need to provide carbon-free electricity to DAC facilities. Differently, BECCS and CCS in the industrial sector are negatively correlated to DAC and they are deployed to a minor extent in scenarios where Direct Air Capture plants can be installed faster and earlier in time; this can be seen with both mitigation targets, but it results more evident with 1.5°C. Again, the difference is not much in the level at the end of the century, but it is more visible in the diffusion path around mid-century. It should be remembered that in any case BECCS deployment is limited by the biomass potential available.

(a) 2° C scenarios　　　　　　　　　　*(b) 1.5° C scenarios*
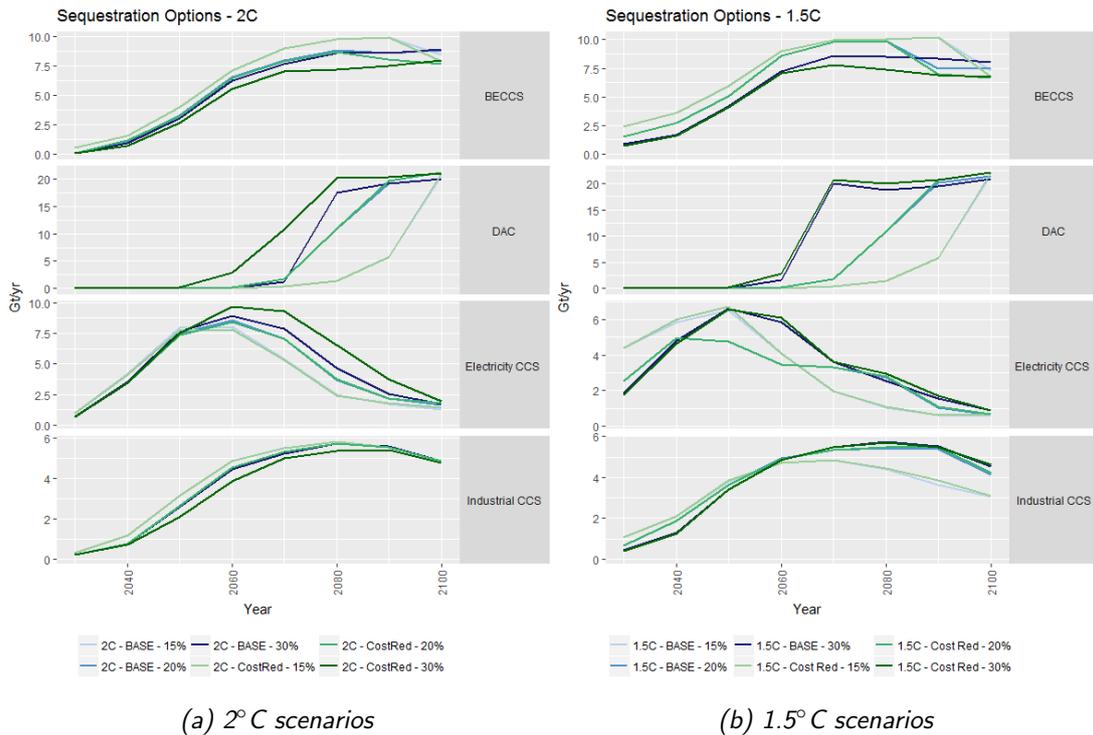
Figure 6.21: Deployment of sequestration options for 2° C and 1.5° C, with different growth rates.

It is interesting to notice that in 1.5°C case the positive correlation between DAC and CCS in the electricity sector disappears in the first half of the century, as more power plants with carbon capture are installed when less DAC capacity can be deployed: indeed, the target results so stringent that all possible mitigation strategies need to be put in place. On the other hand, higher growth rates and more DAC capacity installed earlier in the century result in a reduced role for CCS in the first half of the century with respect to the base case and slightly higher in the second part, so to supply the carbon-free electricity needed by DAC plants once they are deployed massively.

The Energy System

Looking at the renewable capacity, it can be seen that when DAC growth is constrained to a smaller rate, more low-carbon energy sources are needed to decarbonize the energy system, and mainly solar PV is being installed. While with a 2°C target, its deployment is simply shifted earlier in time, with a 1.5°C one there is a significant impact on the overall extent reached, going up to 6000 GW of solar capacity.

Differently, solar thermal capacity does not show a clear trend across different growth rates scenarios, as it is influenced more by DAC2 deployment (to provide the required waste heat) than by the need to supply low-carbon electricity. For 2°C, its capacity shows a spikes in the scenario with a low growth rate and exogenous cost reduction, corresponding to a larger amount of DAC2 plants installed. On the other hand, with 1.5°C, the largest deployment takes place when a high growth rate is applied to DAC: once again, this is the case corresponding to a larger capacity of amine-based plants.
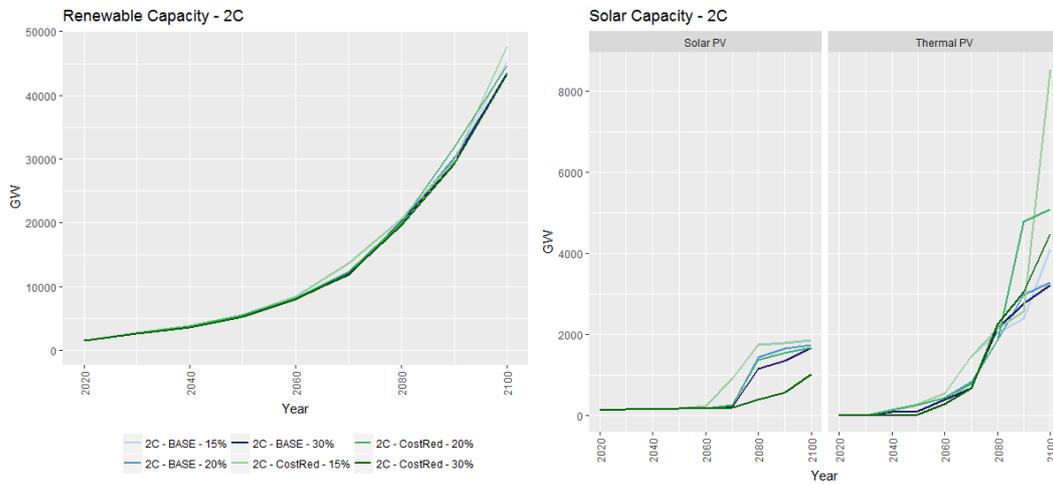
(a) 2°C scenarios
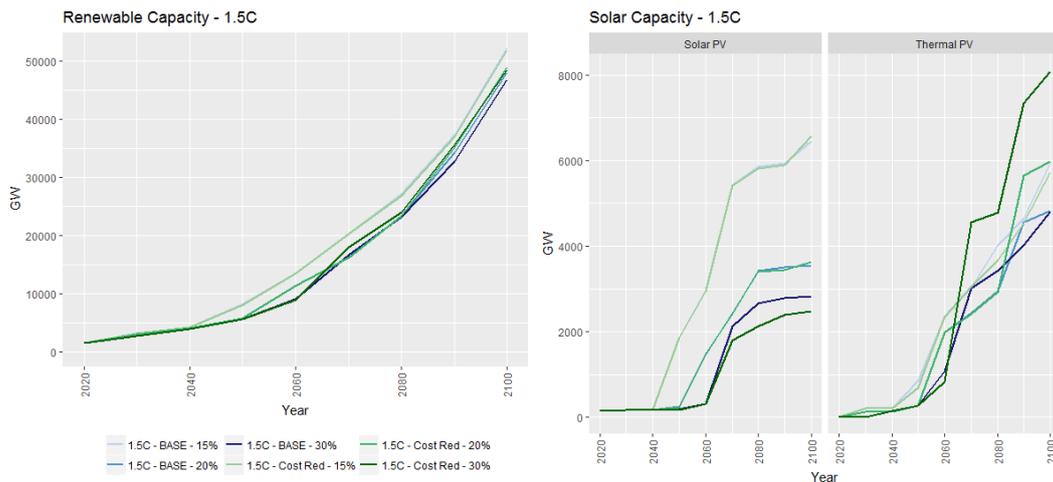
(b) 1.5°C scenarios

Figure 6.22: Renewable capacity installed for 2°C and 1.5°C scenarios, with different growth rates.

Impacts on the electricity mix and the primary energy supply are not much significant within 2°C scenarios: generally, a higher growth rate allows higher share of coal in the electricity mix in 2050 (from 20 to 23%) and consequently a lower need for gas-based power plants, while in 2100 the share of renewables decreases slightly.

Differently, more marked impact can be seen looking at scenarios consistent with a 1.5°C increase in temperature. In particular, it is interesting to notice that a higher growth rate for DAC increases the role of coal in the primary energy supply (see Figure 6.23), allowing coal-based power plants to still play a role in the electricity production up to 2050 with a share around 13% rather than a complete phase-out. On the contrary, lower growth rates require more electricity to be produced by gas-fired plants and overall a higher amount of electricity produced in the mid-century to sustain a massive electrification of the system. The impact on the TPES at the end of the century is less marked than in 2050, but generally it can be seen that with 15% growth rate more renewable sources are needed to meet the (higher) electricity demand compared to the 30% growth case.

Also the impact on the overall system cost is more evident than in 2°C scenarios, leading to an increase of about 7-8% with stringent growth rate, while more rapid growth may reduce it of about 2-3%.
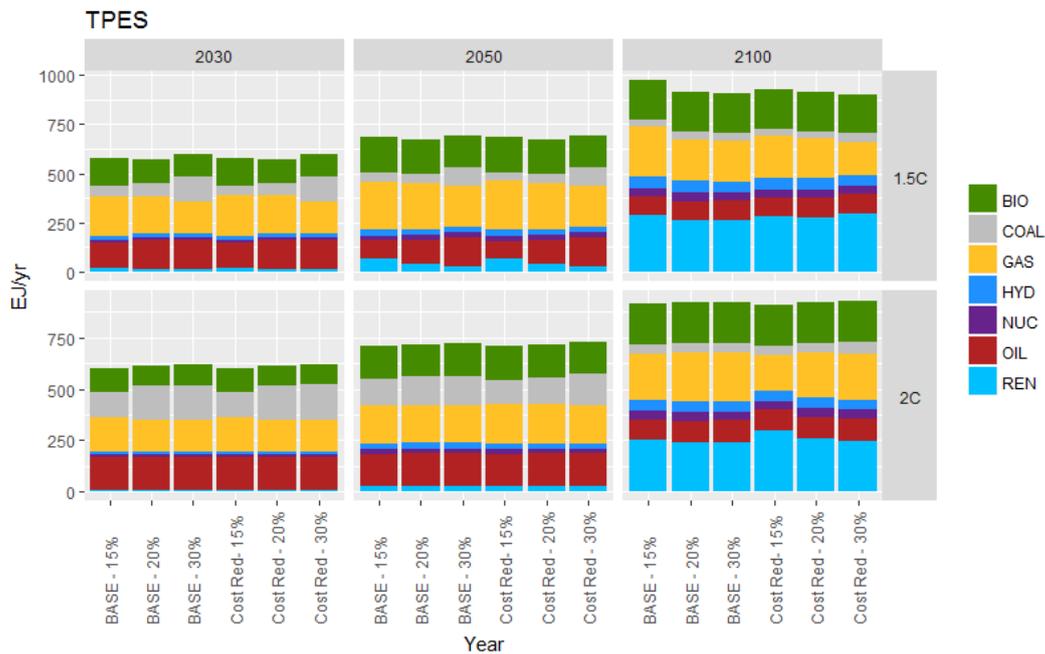
Figure 6.23: Total Primary Energy Supply for 2°C and 1.5°C scenarios, with different growth rates

The trend of sector emissions is quite similar with both mitigation targets, showing a more drastic decarbonization of the transport, residential and industrial sectors when DAC diffusion is limited in the first decades. Once again, the impacts is more marked in 1.5°C scenarios, as it can be seen from Figure 6.24.
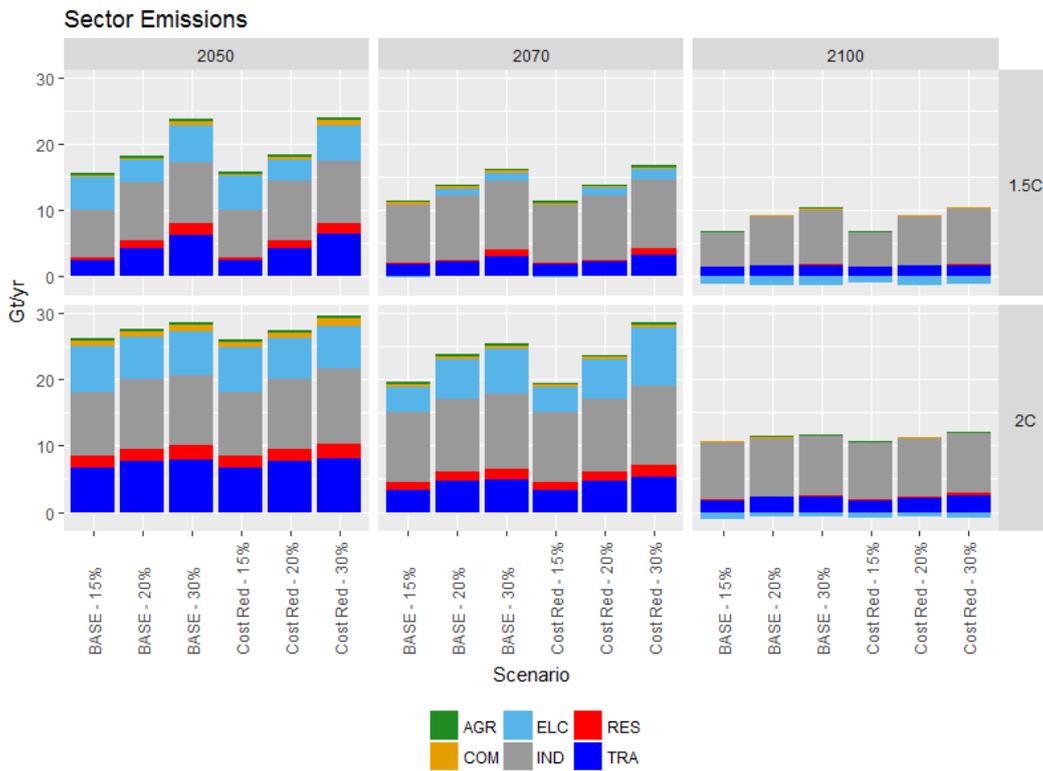
Figure 6.24: Sector emissions for 2°C and 1.5°C scenarios, with different growth rates.

Regional Distribution

As impacts are more evident when a stringent mitigation target is applied, we are going to focus on the regional breakdown only for 1.5°C scenarios, as it is shown in Figure 6.25. Other NET options, namely BECCS and afforestation, are not highly affected on their regional distribution by the different growth rate applied to DAC: in 15% cases, more BECCS is found to be installed in China, USA and other Asian countries (ODA).

The regional distribution of DAC power plants result very interesting across scenarios: indeed, Western Europe (WUE) and Asian countries (ODA) are the main responsible for the increase in DAC capacity with base cost assumptions, similarly to what has been highlighted in previous results analysis, and this increase is mainly related to DAC1 technologies. Differently, with an exogenous cost reduction, the regional breakdown changes, with Mexico (MEX) and mainly Middle East countries (MEA) capturing large cumulative amount of carbon dioxide through amine-based power plants. At the same time the role of Western Europe is much reduced with respect to the base case scenarios with high constant cost.

Considering regional net emissions, it is interesting to notice that China and USA are the main responsible for reduced emissions when more stringent growth rates are allowed for DAC.
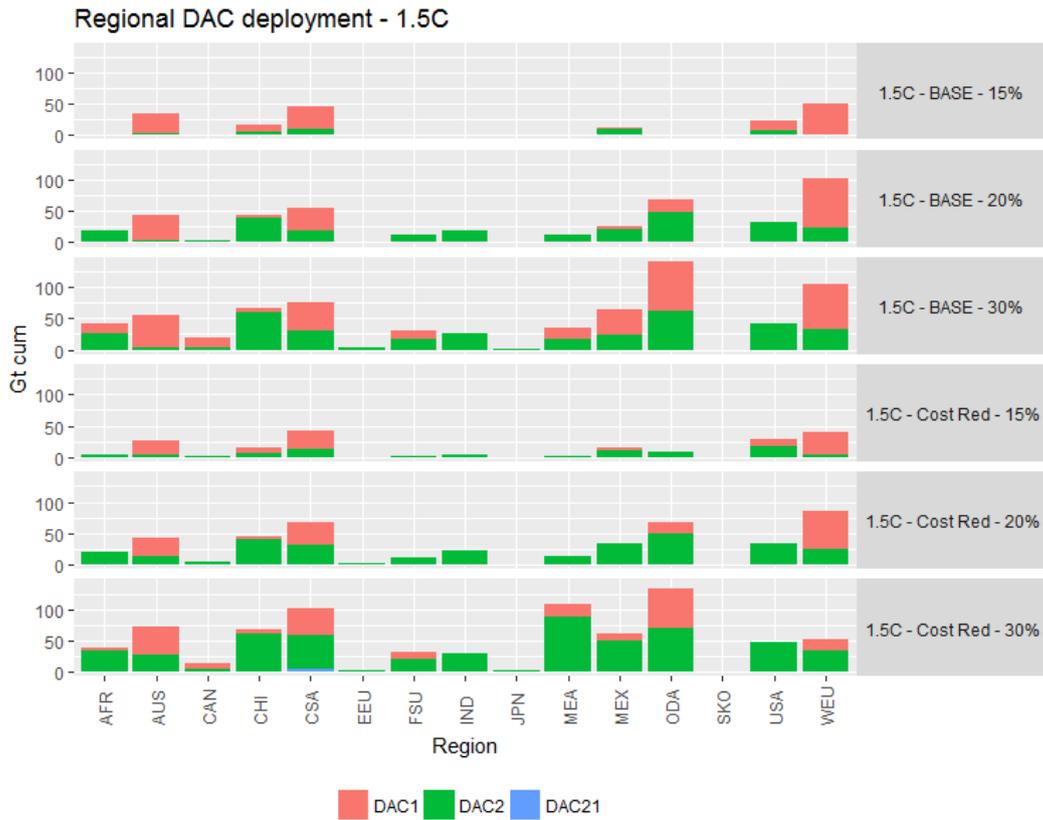
Figure 6.25: Regional breakdown of DAC. Cumulative capture along the century is considered [Gt_{cum}] for 1.5°C scenarios.

6.3.2 Maximum CDR Capacity

Besides the annual growth rate, also different values for the cap applied to Carbon Dioxide Removal options, as the sum of DAC, BECCS and afforestation capture rate-have been explored. According to the literature discussed previously in Section 4.2, negative emissions are limited to 10, 20, 35 and 50 Gt/yr in the model. The results with a 20 Gt cap will not be presented in the charts, as they do not represent an extreme case but simply an intermediate condition between the baseline and the lower bound used to check the robustness of model results. This sensitivity is applied with different assumptions on cost (kept constant in time as in *BASE* scenarios and with ad exogenous reduction as in *Cost Red* ones) and energy (high estimates in the *BASE* scenarios and low in *Low Energy*).

When moving to more stringent mitigation target (1.5 °C), imposing a small cap of CDR removal potential (10 Gt/yr) means that the model is not able to find a solution, therefore these results are not taken into account in the following discussion.

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Scenarios Investigated:

Max CDR Capacity			BASE - constant cost	Cost Reduction	Low Energy
	Low	10 Gt/yr	✓	✓	✓
		20 Gt/yr	not relevant	not relevant	not relevant
2°C	Baseline	35 Gt/yr	✓	✓	✓
	High	50 Gt/yr	✓	✓	✓
	Low	10 Gt/yr	-	-	-
		20 Gt/yr	not relevant	not relevant	not relevant
1.5°C	Baseline	35 Gt/yr	✓	✓	✓
	High	50 Gt/yr	✓	✓	✓

Net Emission Pathways and Carbon Price
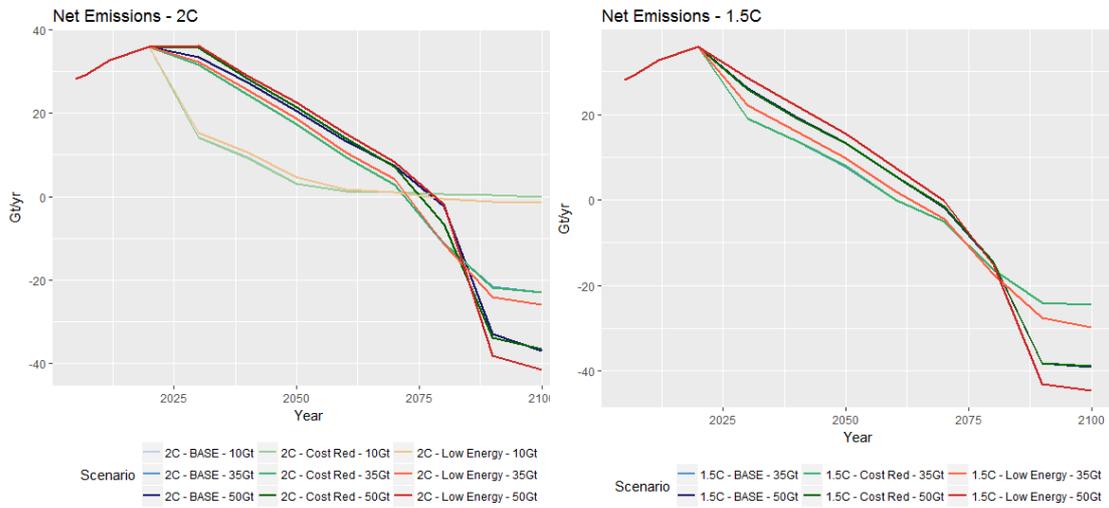
(a) 2°C scenarios   *(b) 1.5°C scenarios*

2°C	Scenario	2020	2030	2040	2050	2060	2070	2080	2090	2100	%
BASE	10 Gt	24	288	470	765	1246	2030	3307	5387	8774	+610%
BASE	35 Gt	24	41	66	107	175	285	465	757	1233	
BASE	50 Gt	24	27	45	73	118	192	314	511	832	- 33%
Cost Red	10 Gt	24	287	467	761	1240	2019	3289	5357	8726	+610%
Cost Red	35 Gt	24	40	66	107	174	284	462	753	1227	
Cost Red	50 Gt	24	24	39	64	105	171	278	453	737	- 40%
Low Energy	10 Gt	24	235	382	622	1014	1651	2690	4381	7136	+580%
Low Energy	35 Gt	24	35	56	92	149	243	397	646	1052	
Low Energy	50 Gt	24	23	37	61	99	162	264	430	700	-33%

1.5 °C	Scenario	2020	2030	2040	2050	2060	2070	2080	2090	2100	%
BASE	35 Gt	24	143	233	380	619	1008	1642	2675	4357	
BASE	50 Gt	24	74	120	195	318	519	845	1376	2241	-49%
Cost Red	35 Gt	24	143	233	379	618	1006	1639	2669	4348	
Cost Red	50 Gt	24	73	119	194	315	514	837	1363	2220	-49%
Low Energy	35 Gt	24	111	181	294	479	780	1271	2079	3372	
Low Energy	50 Gt	24	56	91	148	241	393	641	1043	1700	-50%

Figure 6.26: Net emission pathways and carbon price for 2°C and 1.5°C, with different CDR capacity.

The larger the amount of CDR which is allowed, the more delayed in time mitigation is, so that net emissions remain positive up to 2070 both with 2°C and 1.5°C target, while with a smaller cap they become negative earlier, around 2060, as it can be seen from the charts in Figure 6.26. As expected, the cap applied impacts mainly the extent of negative emissions reached by the end of the time period, moving from around -22/23 Gt/yr to more than 40 Gt/yr when larger capture rate is allowed for NET options. It is interesting to notice that the extent reached is similar with both mitigation targets. Differently, in 2°C scenarios with a 10 Gt/yr cap a huge effort for decarbonizing the system can be found in the first decades, with net emissions remaining close to carbon neutrality from 2060 on. It is interesting to notice that with a stringent cap there is no impact of DAC cost assumptions on the emission pathway.

The carbon price results about 6 times higher when moving from 35 to 10 Gt/yr capture rate with a 2°C target; differently, when increasing the cap to 50 Gt/yr it results about one third and a half, in 2°C and 1.5°C respectively.

DAC Deployment and Other Sequestration Options

Looking at Figure 6.27, it is interesting to notice that with a stringent cap (only 10 Gt/yr allowed in the second half of the century), DAC is not deployed at all in 2°C scenarios, regardless of the cost assumptions, though it is deployed as the only CDR option if energy needs are reduced, replacing also carbon capture through afforestation from 2070 on. This DAC capacity is represented only by strong-base plants (DAC1), as it has been already highlighted that they are the most favored when dealing with *Low Energy* estimates.

2°C Scenario	2060	2070	2080	2090	2100	1.5°C Scenario	2060	2070	2080	2090	2100
BASE - 10 Gt	0.02	0.00	0.00	0.00	0.00	BASE - 10 Gt					
BASE - 35 Gt	0.28	1.75	10.85	19.15	20.04	BASE - 35 Gt	0.28	1.75	10.85	20.28	21.42
BASE - 50 Gt	0.14	0.84	5.23	32.40	34.85	BASE - 50 Gt	0.28	1.75	10.85	33.97	35.63
Cost Red - 10 Gt	0.27	0.14	0.01	0.08	0.50	Cost Red - 10 Gt					
Cost Red - 35 Gt	0.28	1.75	10.85	19.70	21.21	Cost Red - 35 Gt	0.28	1.75	10.85	20.74	22.22
Cost Red - 50 Gt	0.28	1.75	10.85	35.10	35.93	Cost Red - 50 Gt	0.28	1.75	10.85	34.81	35.98
Low Energy - 10 Gt	0.25	1.53	9.45	10.00	10.00	Low Energy - 10 Gt					
Low Energy - 35 Gt	0.28	1.75	10.85	19.77	21.28	Low Energy - 35 Gt	0.28	1.75	10.85	23.26	35.00
Low Energy - 50 Gt	0.15	0.90	5.60	34.67	35.61	Low Energy - 50 Gt	0.28	1.75	10.85	34.81	37.36

Figure 6.27: DAC deployment [Gt/yr] in 2°C and 1.5°C scenarios, with different CDR capacity.

Increasing the cap allowed with respect to the base case from 35 to 50 Gt/yr, the additional CDR capacity is taken mainly by DAC1 when high constant costs are applied, and by DAC2 when the cost is being reduced in time, with both mitigation targets. This confirms how individual DAC technologies are sensitive to energy and cost to different extent.

While 2°C scenarios show higher variability in DAC deployment along the century according to the different cap imposed, with a more stringent mitigation target the deployment of DAC up to 2080 is determined only by the growth constraint applied, being exactly the same across different cost, energy and maximum capacity assumptions. This means that the model needs to deploy as much DAC capacity as possible to meet the budget, regardless of the cost this option may have. From 2090 on scenarios with 35 and 50 Gt/yr cap starts diverging, but again the impact of different cost and energy assumptions is very limited on the overall capacity installed.
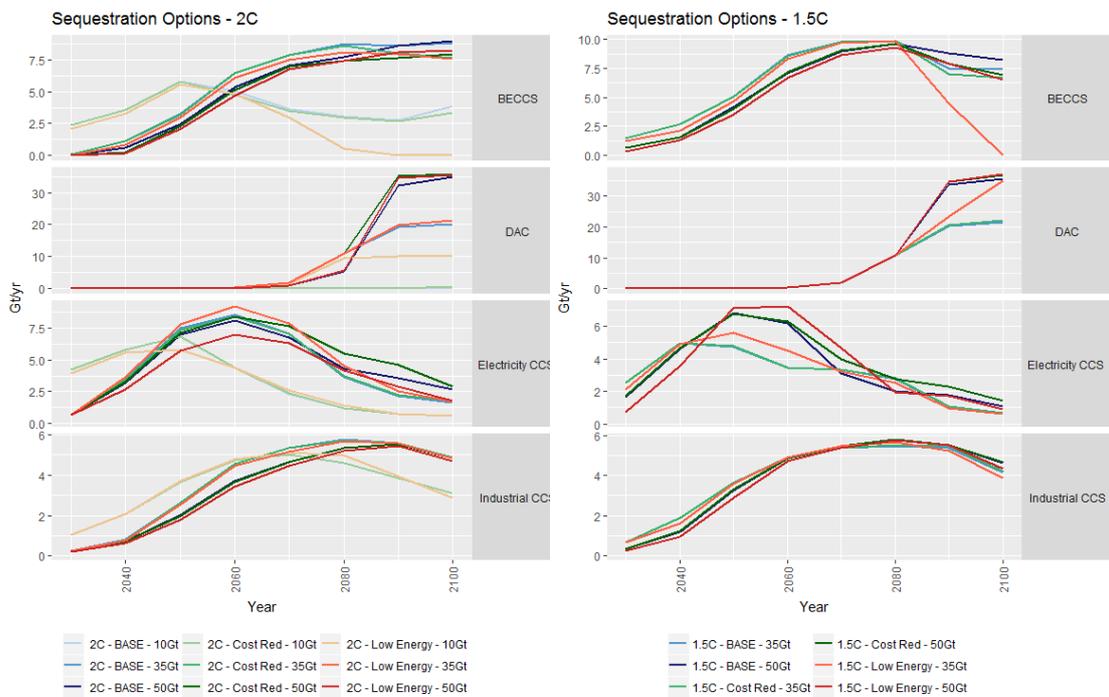
(a) 2°C scenarios

(b) 1.5°C scenarios

Figure 6.28: Deployment of sequestration options in 2°C and 1.5°C, with different CDR capacity.

Considering the other NET options available, it is interesting to notice that, even if the overall cap for NET technologies is increased, BECCS deployment does not change much, and these plants are not able to capture more than 9 Gt/yr of CO_2 across scenarios, given that they are limited by the availability of sustainable bio-energy. It should be noted that the biomass available is not used entirely to feed this type of plants, as part of it can be used to produce heat, but for the model it does not result convenient to deploy BECCS plants to a larger extent[5].

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[5]As a benchmark, it can be noted that also in 1.5°C scenarios without DAC, BECCS plants only

As expected, with a 10 Gt/yr cap applied BECCS results to be largely reduced in the second half of the century, when DAC becomes a competitive technology and the model prefers to achieve negative emissions through direct capture plants rather than employing bio-energy with carbon capture.

Considering sequestration in other sectors, it can be seen from Figure 6.28a that with a stringent cap (10 Gt/yr) CCS in the electricity is able to capture between 4 and 6 Gt/yr in the 2030-2050 period, with an increased role role as a mid-term mitigation strategy. A similar trend can be found also for carbon capture in the industrial sector, but to a minor extent, as more drastic emission reduction is needed in the first decades of the century due to the limited removal that can be achieved through NETs. On the other hand, it is interesting to notice that the capture rate from the electricity sector results higher from 2050 on when a higher cap is applied: this can be explained with the higher amount of electricity required by DAC plants and at the same time the reduced need for a drastic decarbonization of the energy-intensive sectors. While with a 2°C target, CCS deployment with *Low Energy* assumptions result lower when a higher cap is applied to CDR options, with a more stringent target it is the other way round: as the capture rate is similar with both mitigation budgets, this difference can be related to the fact that with a less stringent target DAC2 are deployed when moving from 35 to 50 Gt/yr, while in the other case only DAC1 are installed. This suggests that strong base plants, requiring more electricity, are driving the installation of power plants with CCS to provide this energy input.
Almost no difference can be found in industrial CCS with a 1.5°C target.

The Energy System

The impact on the energy system can be seen from Figure 6.29 below, looking at the changes in TPES across scenarios. It is interesting to notice that the larger the deployment of NET options (and in particular of DAC plants, given that the capture rate of BECCS is not much influenced by the cap applied, as discussed before), the larger the amount of primary energy needed by the system: this is caused mainly by the substantial energy input to operate DAC plants, with the difference being more relevant in the last decades of the century. Generally, it can be seen that the larger deployment of negative emissions allows fossil fuels to cover a higher share of the TPES, with coal being increased up to 2050 and gas in the last part of the century.

It is interesting to notice that, differently from TPES that shows a steadily growth along the century when higher capacity is allowed, the electricity production is higher in 2050 when a more stringent cap on CDR removal is applied (10 Gt/yr) in 2°C scenarios, probably due to the electrification of energy intensive sectors - transport and industry - required to meet the target with a reduced role of negative emissions. The electric demand is met relying more on gas-based power plants than in the baseline (from 6% to more than 30% share), with a complete phase-out from coal.

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capture up to 10 Gt/yr
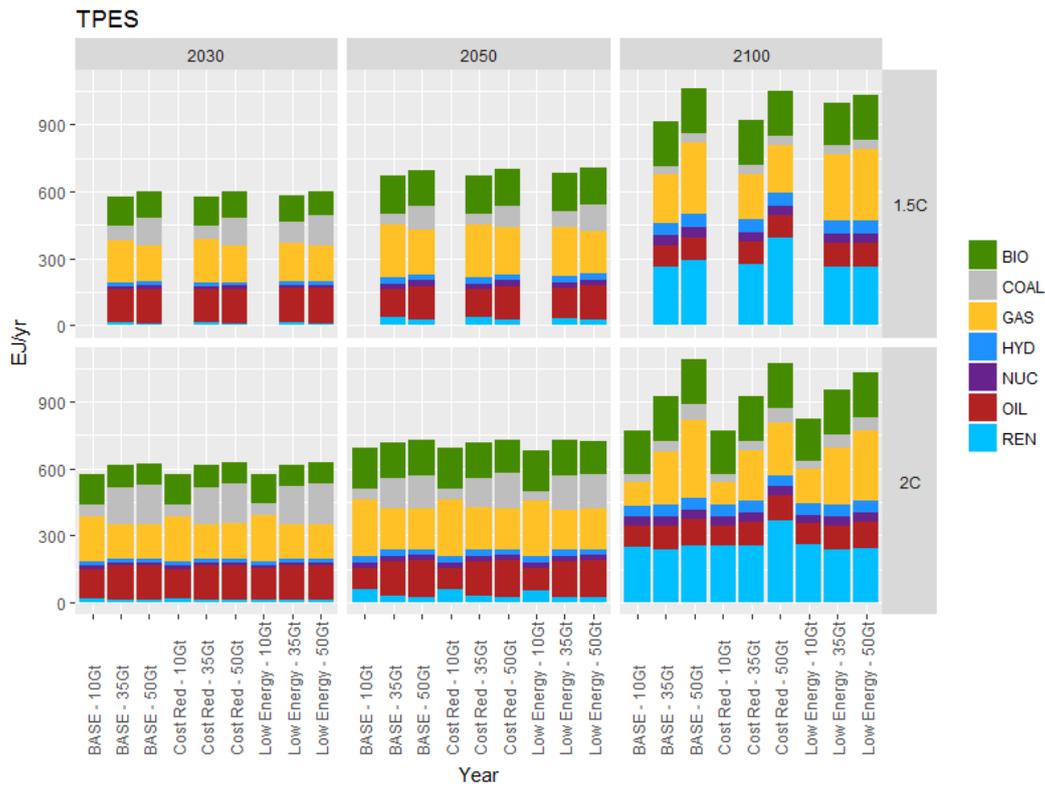
Figure 6.29: Total Primary Energy Supply for 2°C and 1.5°C scenarios, with different CDR capacity.

Differently, in 2100 the electric production result higher in scenarios with a bigger role of DAC (35 and 50 Gt/yr cap), given that DAC plants require between 45 and 50 EJ/yr of electricity in the last decades, representing about 12% of total production.

In 2°C scenarios, a higher NET capacity allows coal to still play a relevant role in the electricity mix in 2050, even with a stringent mitigation target. Moving to 1.5°C scenarios, it is interesting to notice that with 50 Gt/yr of CDR capture rate, electricity production can still rely on coal-based power plants in 2050, delaying their complete phase-out.

Looking at the renewable capacity installed, 10Gt-cap scenarios show an increase in solar PV required to decarbonize the electricity production, while the huge increase in solar thermal is related more to the provision of waste heat to fuel DAC2 power plants[6].

Considering sector emissions, it can be seen that the larger the removal capacity achieved through negative emission technology, the higher residual emissions are, as expected. In particular, this is more relevant for 2°C scenarios, in the transport and the residential sector, while with a more stringent target drastic reduction are needed anyway to remains within the budget allowed.

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[6]indeed, in the *Exogenous cost reduction* scenario with 50 Gt/yr as cap, 85% of waste heat for DAC2 plants come from solar and nuclear plants
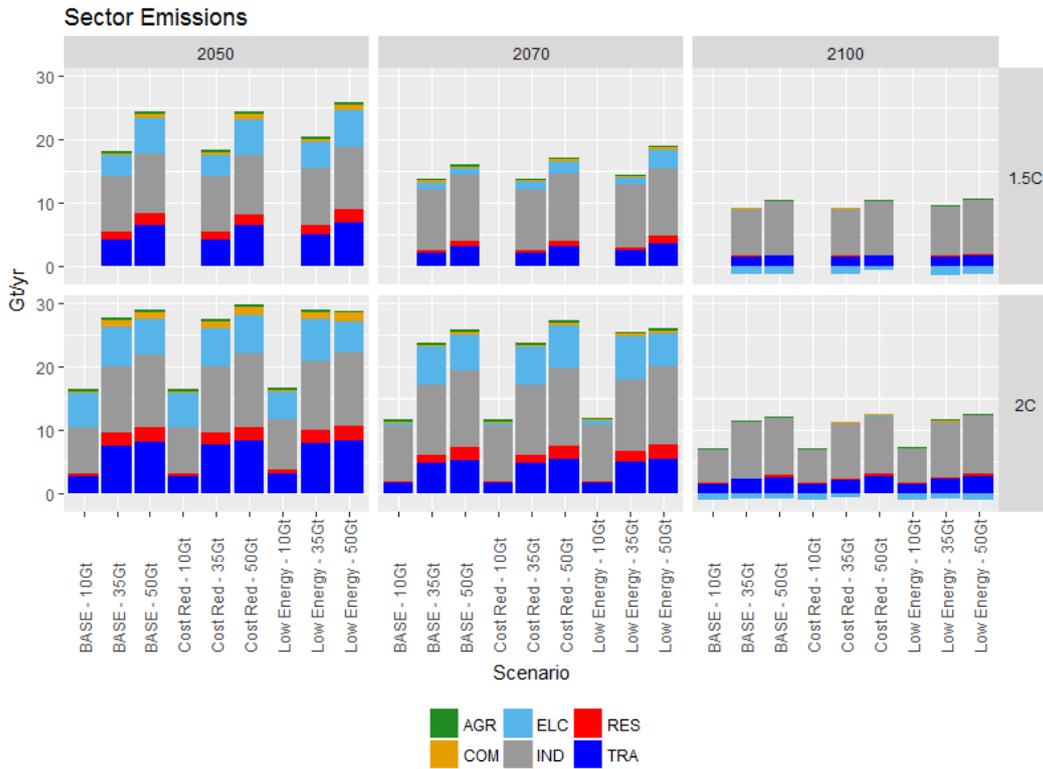
Figure 6.30: Sector emissions for 2° C and 1.5° C scenarios, with different CDR capacity.

6.4 Sensitivity on Storage Potential

Three different scenarios for storage availability have been explored: the baseline, with a very high potential around 9000 cumulative Gton at global level, and two different estimates according to Hendriks [89], namely the *High* one with storage site able to sequester around 5000 Gt, and the *Best* one, with a capacity around 1500 Gt (see Figure 6.31 below). It is interesting to notice that the difference is not only in the overall extent of sequestration resources, but also in their regional allocation, with Hendriks assigning more storage to China and Middle East countries and less to the rest of Asia and to USA compared to the initial assumptions implemented in TIAM. Note that the *Low* estimate by Hendriks, with only 500 Gt available to store carbon dioxide, results too stringent for the model and causes infeasibility, mainly with a 1.5°C target: therefore such scenarios have not be included in the following discussion.

The impact of reduced storage is explored both in scenarios where only DAC is available, to see how/whether it influences its deployment, with a focus at regional level, and with a full NET portfolio, so to investigate the competition with BECCS for the use of storage resources. Given that meeting a 1.5°C target with only DAC technology available appears to be infeasible for the model, leading to a very high carbon price above 200 000 $/ton, sensitivity on storage potential was not considered for this case, but only when the full portfolio of NET technology options is available.

Scenarios Investigated:

	Storage	Availability	Full NET portfolio	Only DAC
	Baseline		✓	✓
2°C	Hendriks	High	✓	✓
	Hendriks	Median	✓	✓
	Hendriks	Low	-	-
	Baseline		✓	-
1.5°C	Hendriks	High	✓	-
	Hendriks	Median	✓	-
	Hendriks	Low	-	-
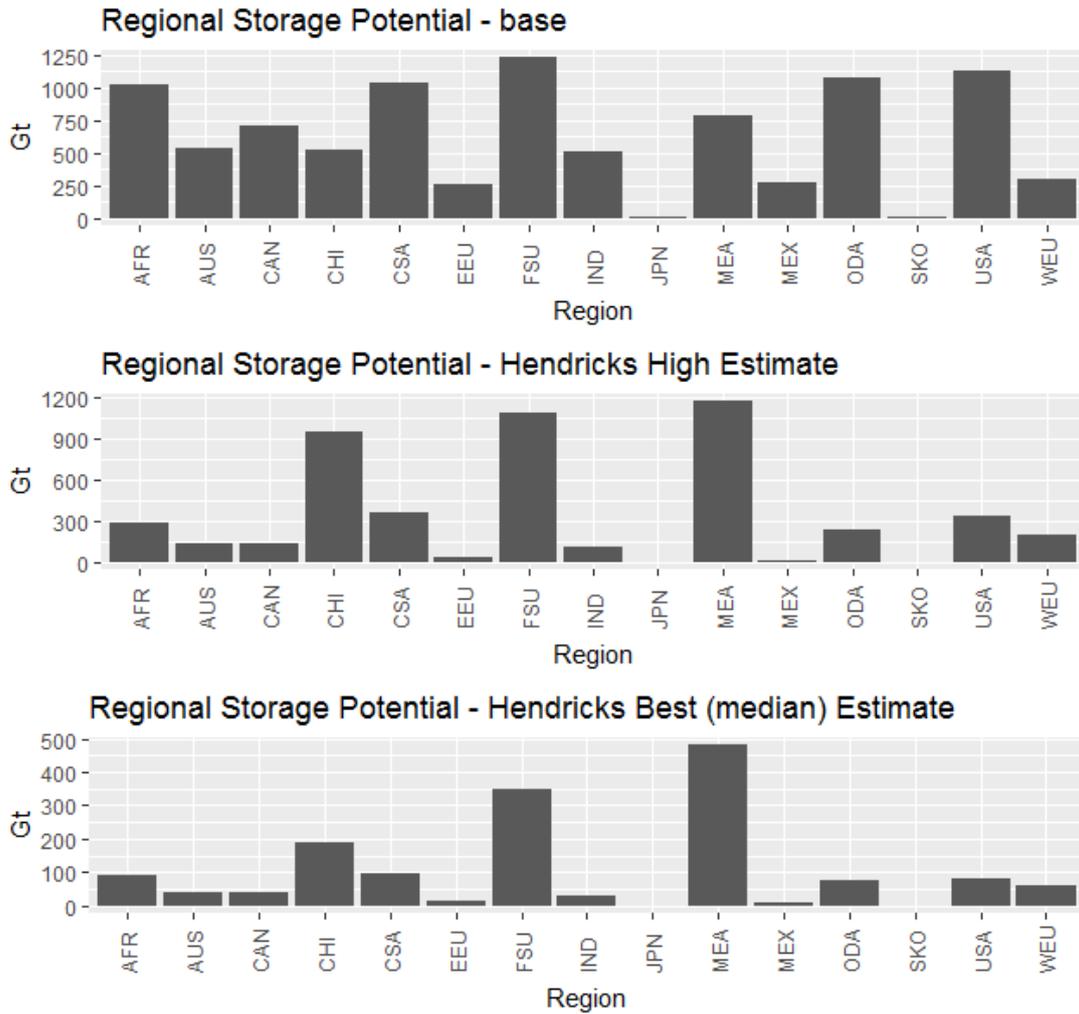
Figure 6.31: Cumulative regional storage availability, according to the different scenarios considered.

In 2°C scenarios, limited storage availability results binding already with *High Storage* estimates in Mexico, China and Western Europe, with a higher marginal value for the constraint in Japan related to Deep Saline Aquifer resource. In 1.5°C scenarios, similar types of storage resources result critical as in the 2°C ones, with more binding

constraints to be found in Australia, Eastern Europe, India and Mexico than before. Limitation are related mainly to coal Bed Methane sites (Australia, Eastern Europe and India), Deep Saline Aquifer, and EOR sites in Western Europe[7].

Note that no significant impact of transport cost can be seen even when there is a reduced storage potential available.

Net Emission and Carbon Price

The net emission pathway and the carbon price do not result to be much influenced by the storage availability, with some differences emerging only when moving to the *Median* estimates by Hendricks: indeed, the marginal abatement cost is about 30% higher than the base case, while applying the high estimates by Hendricks (which are still around half of the initial potential) it only increases by 4%. In scenarios with only DAC the difference in net emission is almost negligible, while it is more visible in the case with a full NET portfolio. With 1.5°C target, the impact net emission pathways is even smaller than before, but the carbon price increases of about 70%.

Nevertheless, limited storage potential (both *High* and *Median* estimates, as the regional distribution of storage is shifted with respect to the base assumptions) do have a huge impact on the overall system cost at the end of the century, as all storage sites need to be used to accommodate the captured CO_2, even the most expensive options.

DAC Deployment and Other Sequestration Options

Generally, it can be said that with reduced storage availability, DAC is given priority on other sequestration options, as the extent of its deployment is not influenced much, while carbon capture in other sectors are largely reduced. Moreover, there is not a large difference when moving from base to *High* storage estimates by Hendricks, meaning that up to 5000 cumulative Gt of CO_2 sequestered this is not a limiting factor, while reducing the potential to about 1500 Gt creates some dynamics in the way sequestration technologies are deployed around the world.

When only DAC is available as a NET technology in 2°C scenarios, its overall deployment is not influenced by the limited availability of storage, being still able to capture 35 Gt/yr from 2090 on, but the limited storage availability do have an impact on the choice of DAC technologies to be deployed. Differently, with a full NET portfolio available, a limited storage availability (*Hendriks - Median*) seems to favor DAC on other sequestration options, as it captures 5 Gt/yr more than in the other cases. This can be clearly seen looking both at Figure 6.32 and 6.33.

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[7]It is interesting to note that this is one of the most abundant storage resource available, but also one of the most favored

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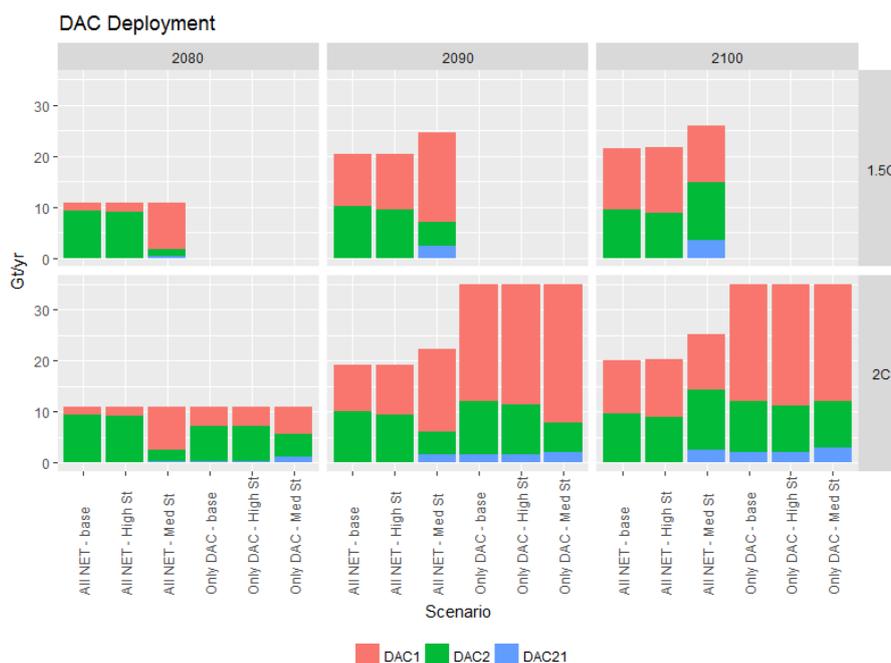
Figure 6.32: DAC deployment [Gt/yr] for 2°C and 1.5C, with different storage potential.

Overall, both for 2°C and 1.5°C, it can be seen that from 2080 on DAC1 is favored over DAC2 when limited storage is available, while in the very last decade the role is reversed. Indeed, in 2100 DAC2 prevails again when combining a full NET portfolio with a reduced sequestration potential, and even DAC21 starts to play a role capturing around 3 Gt/yr, while in previous scenarios it was hardly deployed at all. This could be explained with a greater availability of heat at low price in these scenarios: as the main route for producing low-carbon heat at the end of the century is by using biomass in CHP plants, the reduced role for bio-energy in the electricity sector due to limited storage capacity is freeing part of the biomass to be used for heat production at cheap price so to fuel DAC plants based on solid amine-modified sorbents.

On the other hand, in the mid century a larger availability of gas at low price can be used by DAC1 plants, given that the lower the storage potential and the amount of sequestration that could be deployed, the higher the share of renewables in the energy mix and the lower the amount of gas-based power plants (see Figure 6.34).

Considering other sequestration options, Figure 6.33 clearly shows that with reduced storage availability, priority is given to DAC plants whose role cannot be substituted by other technologies, resulting in less BECCS and CCS in both electricity and industrial sector. Similar trends can be found with both mitigation targets. As expected, afforestation is not influenced at all as it does not require geological sequestration sites to store the CO_2, being a natural sink.

The amount of electricity CCS appears to be drastically reduced during mid-century of 2-3 Gt/yr, having an impact also on the energy mix, as it can be seen in next paragraph. A similar effect can be seen also on industrial CCS but to a minor extent.

(a) 2° C scenarios   (b) 1.5° C scenarios
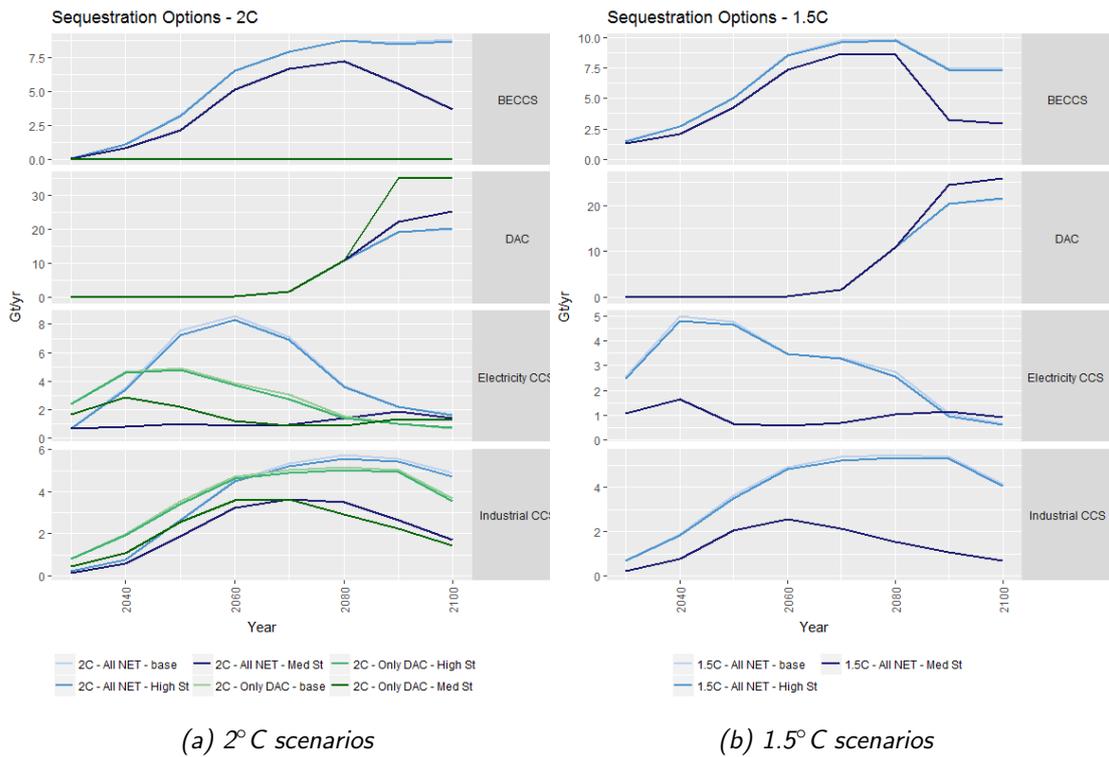
Figure 6.33: Deployment of sequestration options for 2° C and 1.5° C, with different storage potential.

The Energy System

The limited availability of storage do have an impact in the electricity sector, as more renewable capacity (mainly solar PV) needs to be installed to counterbalance the reduced rate of carbon capture by different sequestration technologies throughout the century. This increase is significant for 2°C scenarios in the case with only DAC available, as it is also coupled with an increase in DAC1 plants capacity. Indeed, it has been noted also before that there is a correlation between DAC1 plants and renewable capacity, mainly with solar PV as it is the most "flexible" and cheap source that can be installed by the model, while wind is constrained by a stringent growth rate and nuclear by its large scale. Differently, DAC2 is more related to solar thermal plants that can provide both electricity and waste heat. This can be explained with the fact that DAC1 plants are still associated with a small amount of residual emissions, i.e. captured CO_2 does not correspond with avoided CO_2 as burning natural gas still emit a small amount in the atmosphere, given that the CCS capture efficiency is not equal to 100% (Remember only 95% capture is applied to this plant, as discussed in Section 4.1).

Lower storage potential is also reflected in the electricity mix with a more urgent need for decarbonization earlier in the century: indeed, in 2050 almost no coal is present when the full portfolio of NET is available and a 2°C target is applied, while with 1.5°C the same applies to gas-based electricity production, favoring solar instead.

In 2100 the impact is even more significant, with solar increasing from 13 to 20%, and biomass-based energy decreasing from 7 to 4%. in 2°C scenarios. Moreover, the overall electricity demand results higher in the last part of the century when a limited storage availability is applied, across all mitigation targets, due to a higher electrification needed in specific sectors. A similar behavior is reflected in the TPES as well.
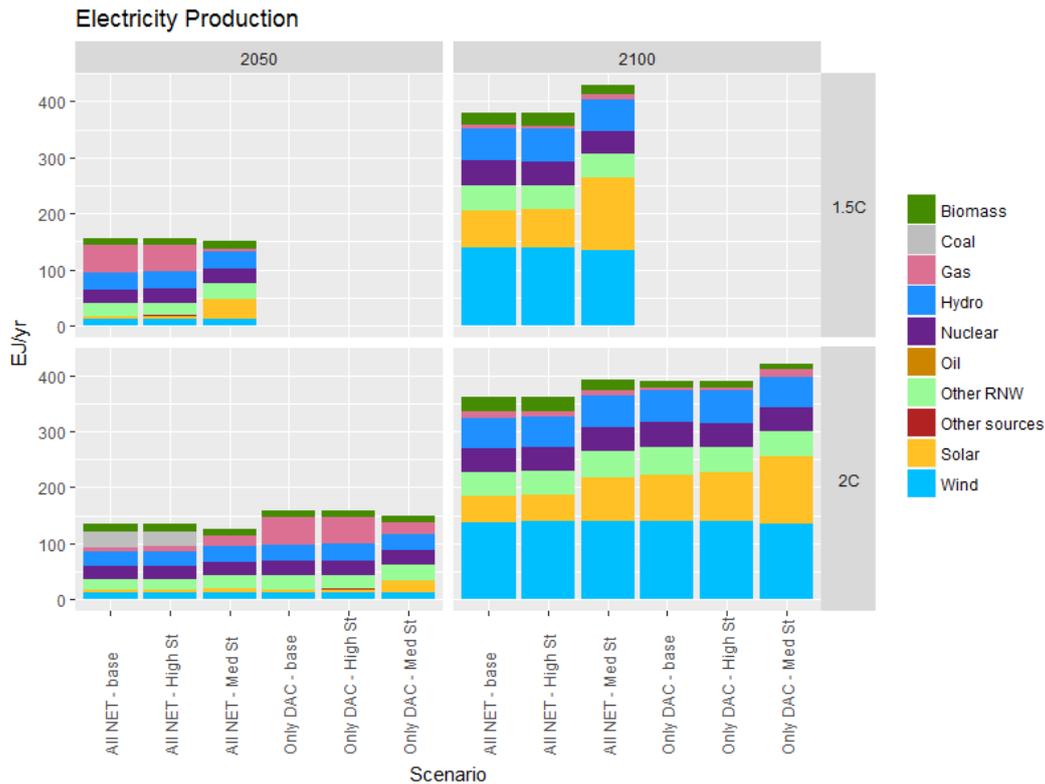
Figure 6.34: Electricity mix for 2° C and 1.5° C scenarios, with different storage potential.

As net emissions do not change much across scenarios, this significant decarbonization of the electricity (and energy) sector can be explained with the reduced deployment of industrial and electricity CCS, as in these sectors carbon capture could be replaced with other mitigation and decarbonization strategies (e.g. low-carbon renewables, electrification of industrial processes), while DAC plants are prioritized to have access to the limited storage availability to offset emissions coming from transport and industry. This is confirmed looking at sector emissions in Figure 6.35: transport and industry emissions does not change much across scenarios, while in the energy sector they become negative already in 2050. Indeed, the lower the storage available, the more emissions need to be reduced early in the century (2020-2040), mainly in the energy sector due to the strong reduction in CCS deployment.
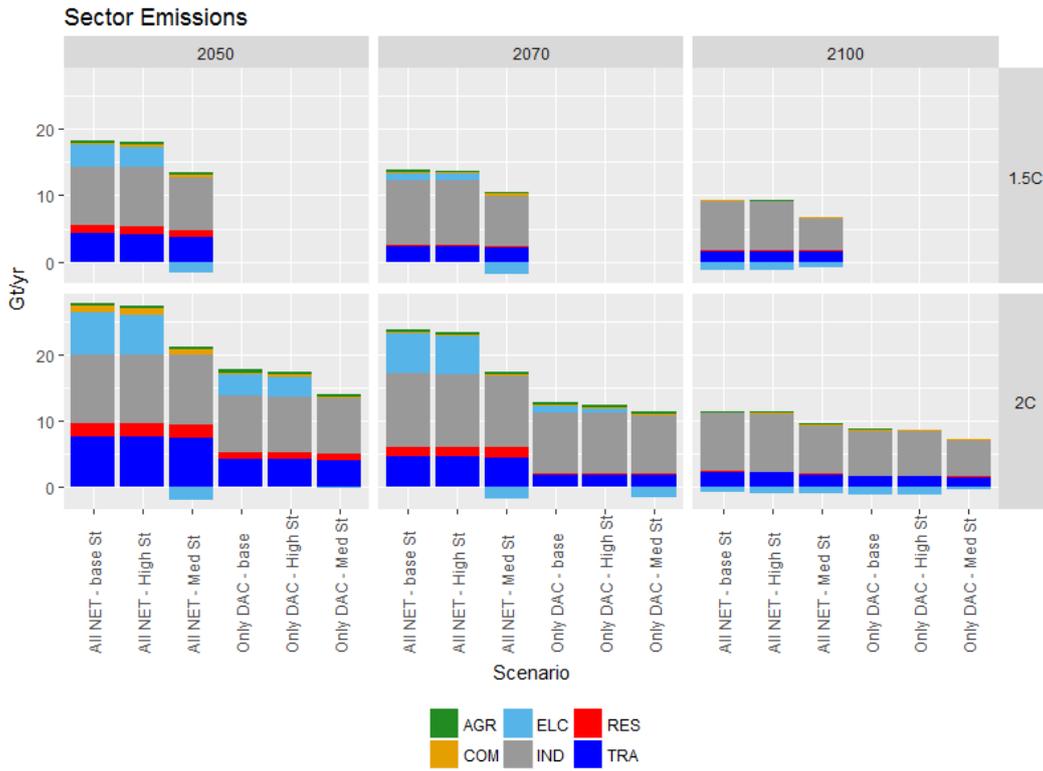
Figure 6.35: Sector emissions for 2° C and 1.5° C scenarios, with different storage potential.

6.4.1 Regional Breakdown

When applying different estimates for storage availability, the share of the overall potential that is assigned to every single region may change a lot across scenarios. As it can be seen from Figure 6.36, Middle East countries and Former Soviet Union represent the majority of the global storage available in both scenarios based on Hendricks estimates, together with China, while in the initial one the total capacity is distributed more evenly across regions.

The regional distribution of storage sites will influence where CDR plants are being deployed, as there is no possibility to "trade" the sequestered CO_2, the carbon dioxide is sequestered in the same region where it is captured. This is much more evident in the *Median* storage scenario, where most of CDR capacity in installed in the regions that hold majority of storage potential (MEA, FSU and CHI). It is interesting to notice that, while in Russia and Central-South America the NET portfolio remains quite diversified, in Middle Eas t(MEA) only DAC plants are being installed, mainly the ones based on strong base solutions. Moreover, while in Middle East a fraction of the installed capacity is still based on solid amine sorbents, in the Former Soviet Union region it is almost completely based on wet scrubbers of KOH solutions (DAC1). This reflects the high abundance of natural gas resources in these countries at low price that could be used as input for providing energy need to DAC1 plants.
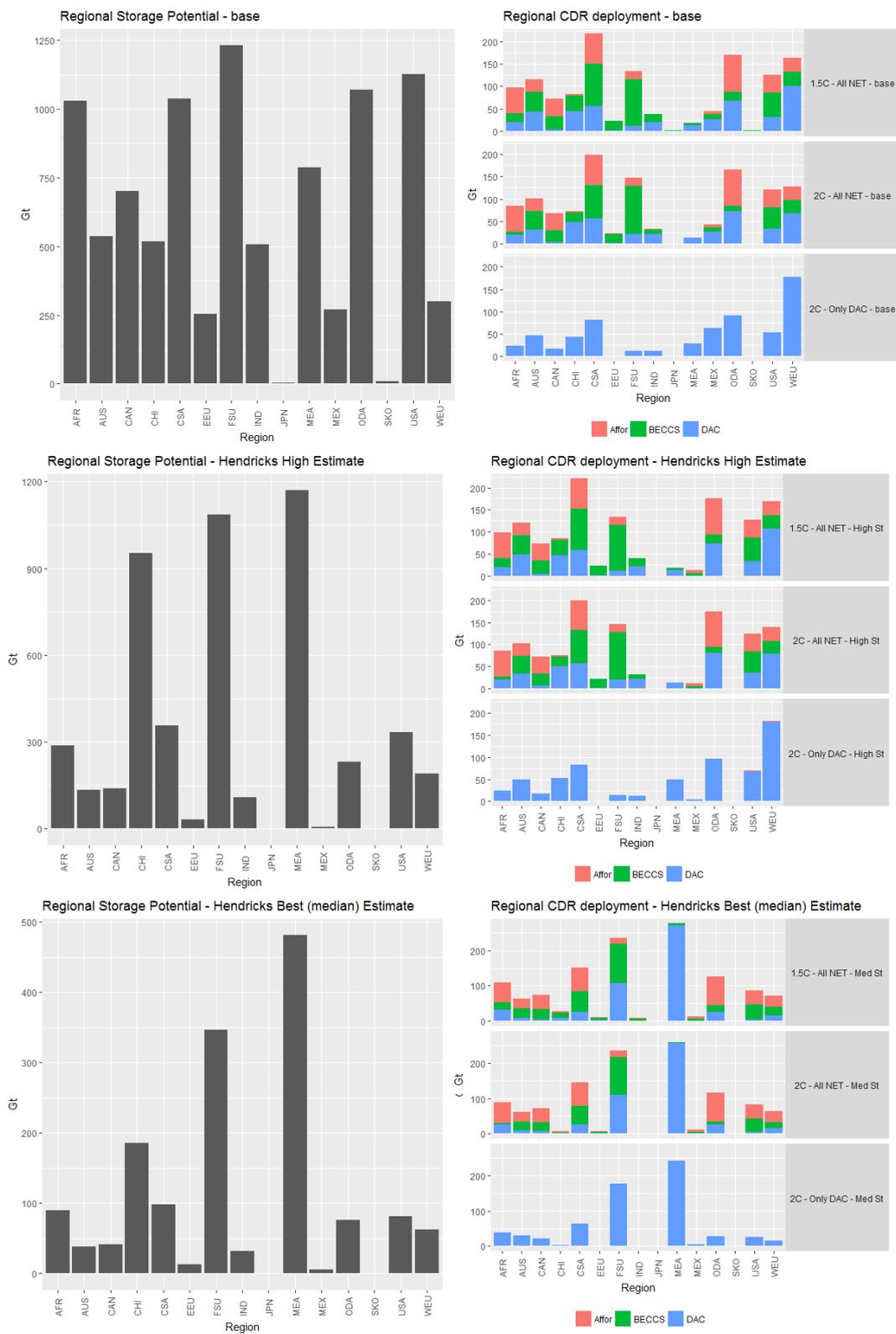
Figure 6.36: Regional breakdown for the storage availability and the corresponding deployment of CDR options, across different storage scenarios, for 2° C and 1.5° C.

Moreover, while in some countries, moving from high to median storage potential estimate, BECCS captured capacity is being replaced by DAC plants (China - CHI - and Russian region - FSU), in most of them also the amount of DAC is reduced as it is concentrated in the few countries that hold consistent storage resources and are characterized by lower commodity price. This means that in the base scenario the regional cost of energy commodities (heat, natural gas and electricity) is not the main factor influencing DAC deployment, while it becomes relevant when storage is a limiting factor, together with the availability of storage itself. Indeed, it should be noted that the amount of CO_2 sequestered by DAC/CDR in China does not increase much with limited storage availability, even if it is one of the region that holds a significant fraction of the global potential, as it does not result economically convenient due to the cost of energy in this country (Note that in China mainly DAC2/21 is installed).

6.4.2 Global Storage Availability

Given that the regional availability of storage result to be the main factor influencing the regional distribution of CDR options, I was interested in investigating which regions would install more DAC and BECCS based on other influencing factors, as commodity price. Therefore, the same total storage potential of previous scenarios has been imposed as a global constraint (i.e. as a sum across all regions, instead of being defined as a regional bound), to understand the extent to which regional or global storage availability influences the distribution of DAC across regions.

Scenario		Affor	BECCS	DAC	Total
1.5°C	High Stor	376	481	442	1299
1.5°C	High Stor Global	376	487	439	1302
1.5°C	Initial	376	487	439	1302
1.5°C	Initial Global	376	487	439	1302
1.5°C	Med Stor	376	369	504	1249
1.5°C	Med Stor Global	376	487	437	1300
2°C	High Stor	376	403	423	1202
2°C	High Stor Global	376	406	421	1203
2°C	Initial	376	406	421	1203
2°C	Initial Global	376	406	421	1203
2°C	Med Stor	376	294	478	1148
2°C	Med Stor Global	376	399	420	1195

Figure 6.37: Cumulative deployment of CDR options for 2°C and 1.5°C scenarios, comparing global and regional storage availability.

A difference is visible only with a limited storage availability (*Med Storage*): when the overall cumulative storage resources is about 1500 Gt, its regional allocation becomes a limiting factor for the deployment of CDR options, as the overall cumulative CO_2 captured along the century results to be reduced of about 50 Gt compared to other cases, as it can be seen from table in Figure 6.37. In all other scenarios, it does not really make a difference if this potential is allocated with a specific regional breakdown or on global scale, as CDR is installed where it is more convenient (and needed), not influenced by where storage is available.
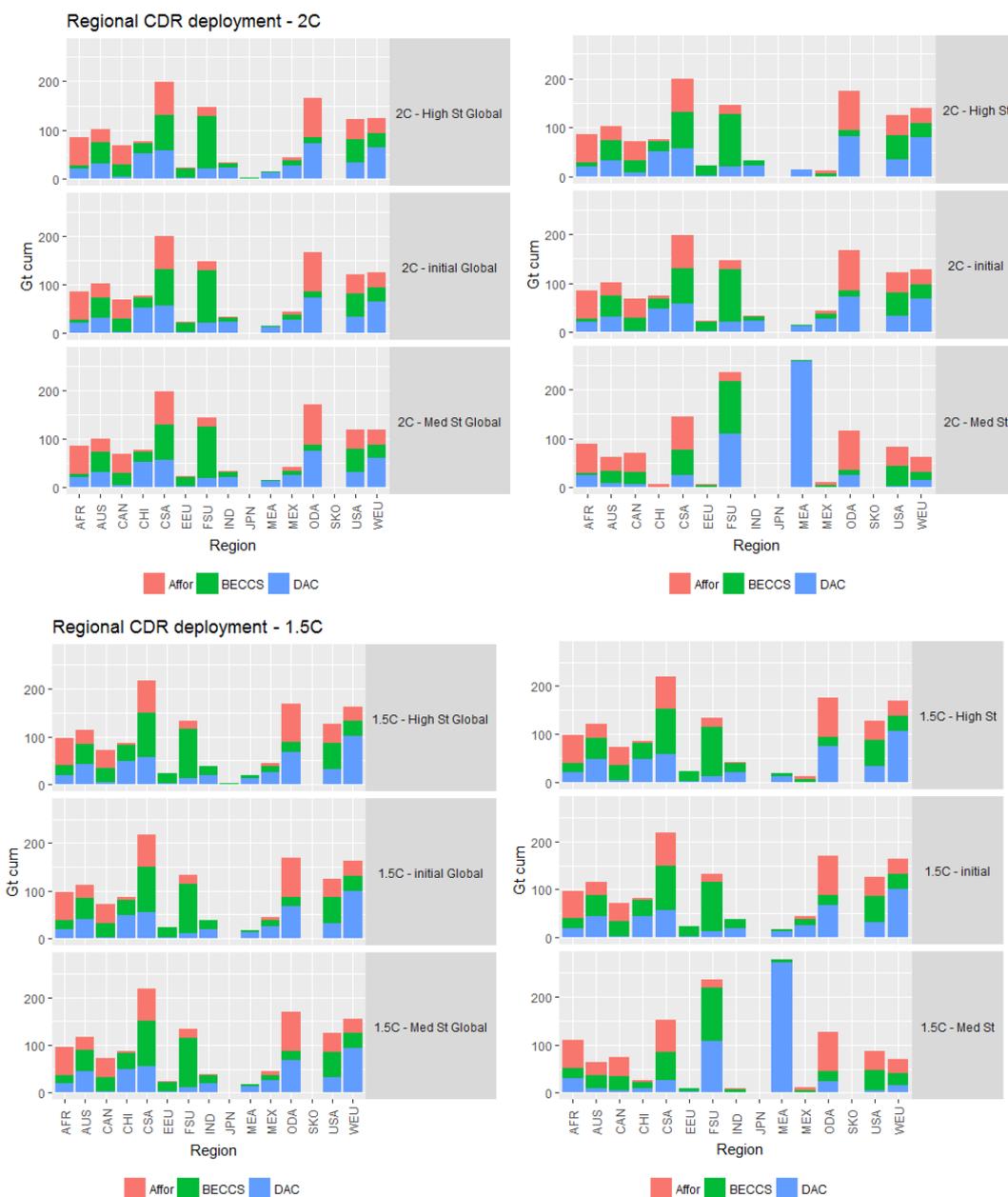
Figure 6.38: Regional breakdown for different sequestration options. Cumulative capture along the century is considered [Gt_{cum}], for 2°C and 1.5°C, with global and regional storage availability.

It is interesting to highlight that the limiting aspect is not the overall amount of storage (even in the initial case, no more than 1300 Gt cumulative are being captured), but how this is distributed among regions, especially when the amount assigned to each of them is reduced as in *Med Storage* scenarios. Indeed, it can be seen that the regional breakdown does not change much when different storage potential are defined on a global level (charts on the left side of Figure 6.38).
With *High estimates*, differences between local and global storage availability are really

small[8], with only Western Europe reducing its cumulative capture by DAC plants and Mexico increasing it, both with 2°C and 1.5°C targets. More significant changes in the regional breakdown can be seen with *Median* estimates, as with storage capacity assigned to each region Middle East Countries and Former Soviet Union (FSU) are installing most of the overall CDR capacity, mainly as DAC, while moving to a global capacity the amount of DAC installed in Middle East countries and FSU is being dramatically reduced, increasing it in Asia (CHI and ODA), Western Europe (WEU) and USA, as well as South America and Australia. (look at Figure 6.38).

6.5   Sensitivity on Biomass Potential

As discussed previously in Section 4.4.3, one important parameter that should be taken into account when investigating the potential role of negative emission technologies as a mitigation strategy is the amount of bioenergy that can be used to sustain the deployment of these options without entering in competition with other sustainable goals, such as food and water supply. Therefore, sensitivity has been done also on this parameter, reducing the amount of bioenergy available in the second half of the century from the initial value of 200 EJ/yr to 120, 90 and 60 EJ/yr, according to different estimates found in the literature.

Note that all scenarios with bioenergy limited to 60 EJ/yr, corresponding to the current level, and 90 EJ/yr show infeasibility in term of dummy energy imports. While in the case of 60 EJ/yr the scale of infeasibility is really high throughout the century, in the latter case this is limited, therefore I am including results with 90 EJ/yr cap, with some caution in making conclusions out of them.

At first, the impact of limited bioenergy potential is investigated only on DAC technologies: as already discussed, a stringent carbon budget consistent with 1.5°C temperature increase is not likely to be be met relying only on Direct Air Capture, as this would result in extremely high system costs, therefore these infeasible scenarios are not included in the following discussion, and I will focus only on 2°C scenarios. Then, the impact of bioenergy potential is investigated considering a full NET portfolio for both mitigation targets.

Scenarios Investigated:

	Biomass	Availability	Full NET portfolio	Only DAC
	Baseline	200 EJ/yr	✓	✓
2°C	High	120 EJ/yr	✓	✓
	Medium	90 EJ/yr	✓	✓
	Low	60 EJ/yr	-	-
	Baseline	200 EJ/yr	✓	-
1.5°C	High	120 EJ/yr	✓	-
	Medium	90 EJ/yr	✓	-
	Low	60 EJ/yr	-	-

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[8]Note that in initial case the availability is so high for each region that distributing it on a global scale does not change the regional breakdown at all

6.5.1 Limited Bioenergy with Only DAC

As expected, the lower the biomass potential, the higher the carbon price and the more renewable capacity that need to be installed to meet the mitigation target imposed. With only DAC available as a carbon removal technology, the impact on the net emission pathway is very limited, also because DAC is not directly related to bioenergy use. Indeed, between 2030 and 2050 emissions are reduced of about 4 Gt/yr and in the same years a higher deployment of CCS can be found both in the electricity and the industrial sector, while from 2060 on they result slightly higher of about 2 Gt/yr. Differently, a larger impact will be noticed a full NET portfolio is considered, including also BECCS (see next Section 6.5.2).

(a) DAC deployment [Gt/yr]   (b) Other sequestration options [Gt/yr]

Figure 6.39: Deployment of DAC and sequestration option for 2°C, with different biomass potential.

Looking at the deployment of DAC as shown in figure 6.39a, when no BECCS or afforestation are available the impact of reduced biomass potential can be seen mainly on amine-based plants: as the heat needed to fuel part of these plants (DAC21) is being produced mainly by bioenergy-based CHP plants, that removal capacity is now achieved relying only on waste heat, that means shifting from DAC21 to DAC2, while DAC1 is almost not influenced at all and the overall capacity installed results exactly the same. On the other hand, limited bioenergy available increases the role of CCS in the power sector in mid century, between 2030 and 2060 (see Figure 6.39b), while increasing the overall electricity production and the share of intermittent renewables in the mix, as bioenergy plants are not deployed. Indeed, both with 120 and 90EJ/yr of primary bioenergy potential, biomass does not play a role in the electricity production.
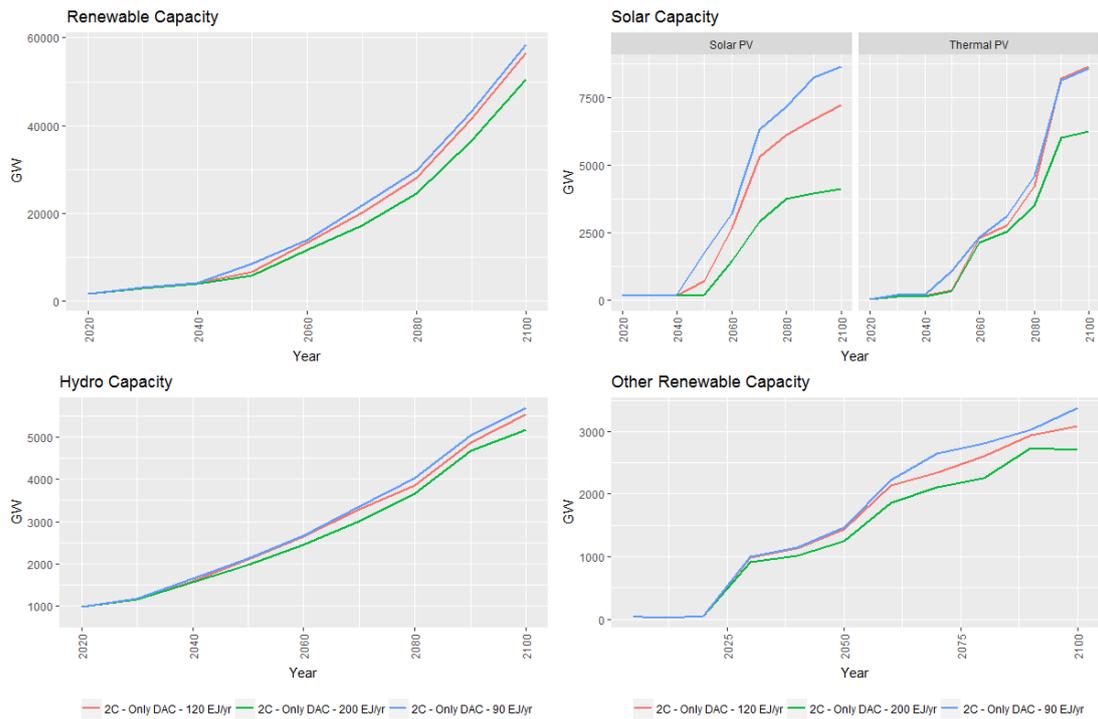
Figure 6.40: Renewable capacity installed for $2°C$, with only DAC and different biomass potential.

A major implication of biomass limitation can be found in the heat production, as in baseline scenarios most of the heat commodity for residential and commercial demand is being produced through bioenergy in CHP plants by the end of the century. When bioenergy is limited to 120 and 90 EJ/year, it can be found that DAC21 plants (that originally account for 65% of heat demand) is drastically reduced, together with the amount of heat needed in commercial sector.

Looking at the flow of biomass commodities summarized in Table 6.1, it can be seen that the ones most affected (as well as the most used) are bioenergy crops (1st generation) and solid biomass (2nd generation of bioenergy), as well as the commodity representing the end-use of biomass in different sectors. Generally this is dramatically reduced in the electricity sector and for the production of biofuels, with a contraction ranging between 60 to 90%.

	COMM	IND	RES	ELC	Biofuel[a]
1st gen Biocrops	-	-	-	-70/-90%	-60/ -75%
2nd gen Biomass	-	-	-	-60/-70%	-35/ -50%
End-use Sectors	-10/-20%	-15/-20%	-25/-30%	-50/-60% [b]	-

Table 6.1: Reduction in primary biomass commodities flow to different sectors, when a limited bioenergy potential is applied, for $2°C$ scenarios with only DAC.

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[a]Both biodiesel and FT synthesis

[b]These are represented by biofuels for electricity sector)

6.5.2 Limited Bioenergy with Full NET Portfolio

At this point, the effect of a limited bioenergy availability is compared when the full portfolio of NET is available, across 2°C and 1.5°C scenarios, therefore including also impacts on the deployment of BECCS.
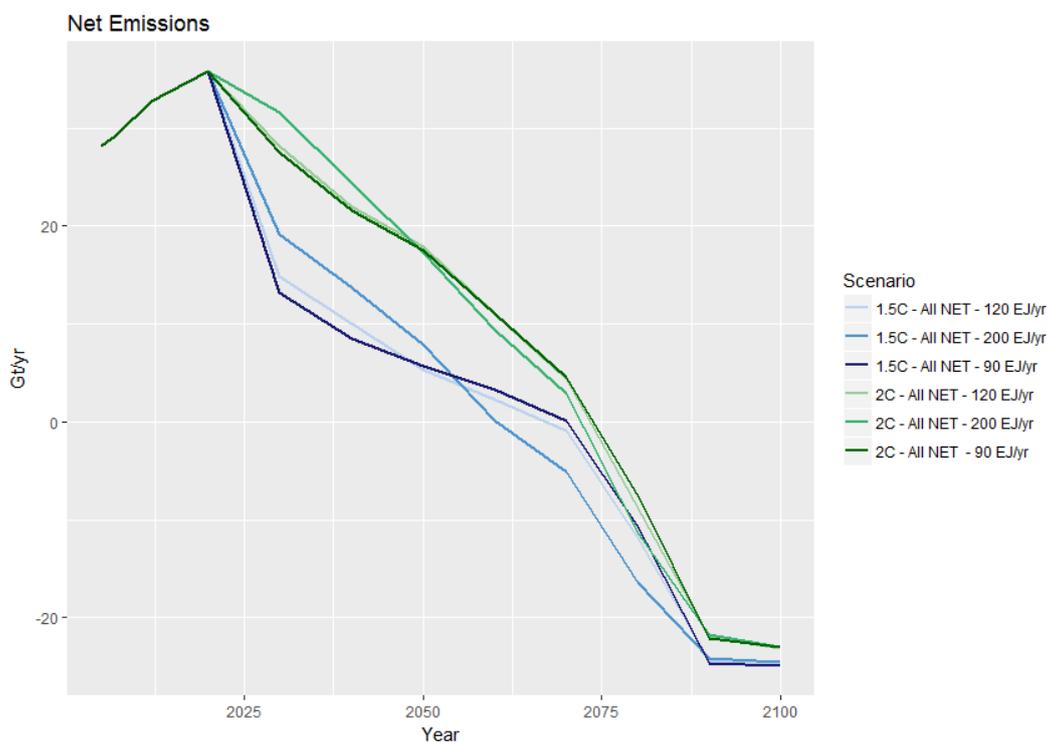
Scenario			2020	2030	2040	2050	2060	2070	2080	2090	2100	%
2°C	Full NET	90 EJ/yr	24	85	139	226	369	600	978	1593	2594	+110%
2°C	Full NET	120 EJ/yr	24	69	113	183	299	487	793	1291	2103	+71%
2°C	Full NET	200 EJ/yr	24	41	66	107	175	285	465	757	1233	
1.5°C	Full NET	90 EJ/yr	24	650	1059	1726	2811	4579	7458	12149	19789	+350%
1.5°C	Full NET	120 EJ/yr	24	358	583	950	1548	2521	4107	6690	10897	+150%
1.5°C	Full NET	200 EJ/yr	24	143	233	380	619	1008	1642	2675	4357	

Figure 6.41: Net emission pathways and carbon price for 2°C and 1.5°C scenarios, with a full NET portfolio and different biomass potential.

Both in 2°C and 1.5°C scenarios, we do have an impact on the net emission pathways along the century, as mitigation starts earlier when limited bioenergy is available, thanks to an increased role of CCS in the electricity sector between 2030 and 2060. This effect can be seen more clearly with a stringent mitigation target, leading to much more drastic emission reduction between 2020 and 2050, with a carbon price more than 3 times higher than the base case.

DAC Deployment and Other Sequestration Options

Generally, it can be seen from Figure 6.42a that the lower the amount of bioenergy available, and thus the deployment of BECCS as a carbon removal option, the

more important the role assigned to DAC plants to achieve the imposed target, increasing its capacity of about 5 Gt/yr. Again, it can be noticed how the limited availability of bioenergy affects mainly DAC1, which is deployed more when BECCS is highly constrained (i.e. with 90 EJ/yr constraint). While with a 2°C target DAC2 deployment is not influenced, showing a marginal increase of strong base solutions plants, with 1.5°C not only the capacity of DAC1 plants results higher, but DAC2 is even reduced.

The impact on other sequestration option, namely CCS in electricity and industrial sector and BECCS, is very similar across both mitigation targets, but more marked in the 1.5°C case. From Figure 6.42b, it can be noticed that while bioenergy with carbon capture is strongly reduced by the limited availability of biomass feedstock, with its capacity being more than halved, CCS in the electricity sector results higher in the first part of the century, then decreasing once DAC is deployed massively. Differently industrial CCS is reduced with a 1.5°C target and a lower bioenergy use.

Scenario			2060	2070	2080	2090	2100
2°C	Full NET	90 EJ/yr	0.28	1.75	10.85	24.96	26.53
2°C	Full NET	120 EJ/yr	0.28	1.75	10.85	23.29	25.00
2°C	Full NET	200 EJ/yr	0.28	1.75	10.85	19.15	20.04
1.5°C	Full NET	90 EJ/yr	0.28	1.75	10.85	26.26	27.44
1.5°C	Full NET	120 EJ/yr	0.28	1.75	10.85	24.57	25.99
1.5°C	Full NET	200 EJ/yr	0.28	1.75	10.85	20.28	21.42
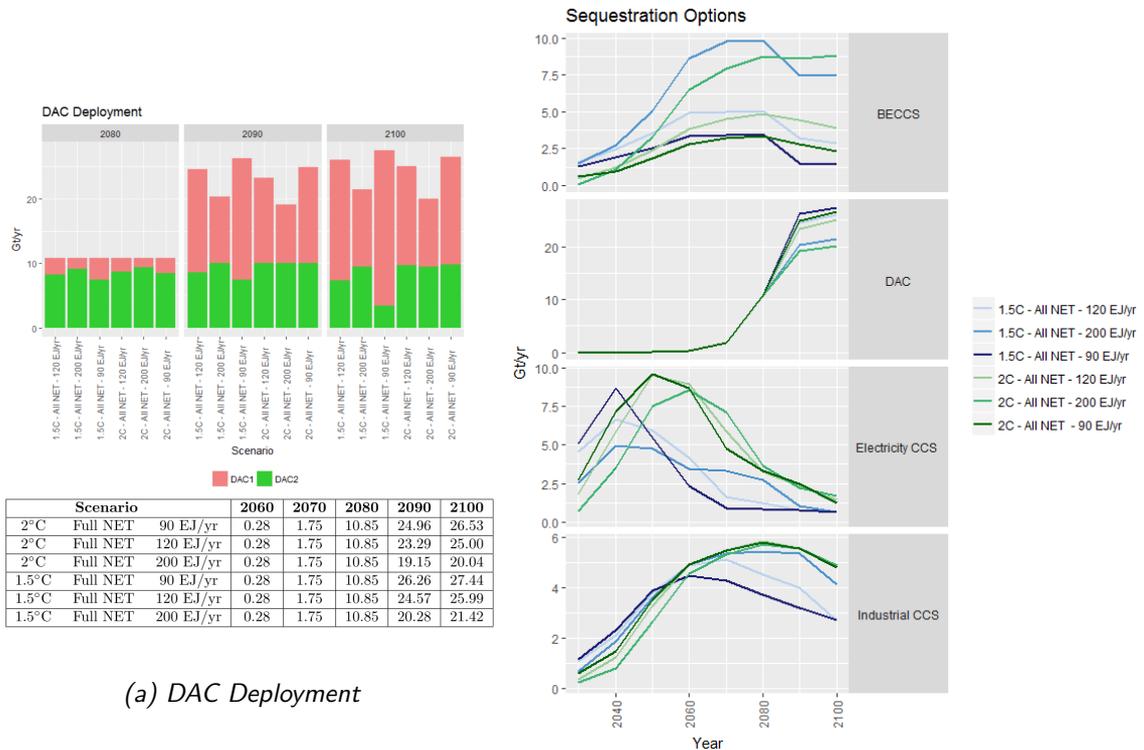
(a) DAC Deployment

(b) Other Sequestration Options

Figure 6.42: Deployment of DAC and other sequestration options for 2°C and 1.5°C, with a full NET portfolio and different biomass potential.

The Energy System

Looking at the electricity mix (Figure 6.43 below), it can be clearly seen that the lower the biomass potential the greater the role for intermittent renewable sources (mainly solar) and for gas in the energy system, with a consequent increase in its total cost.
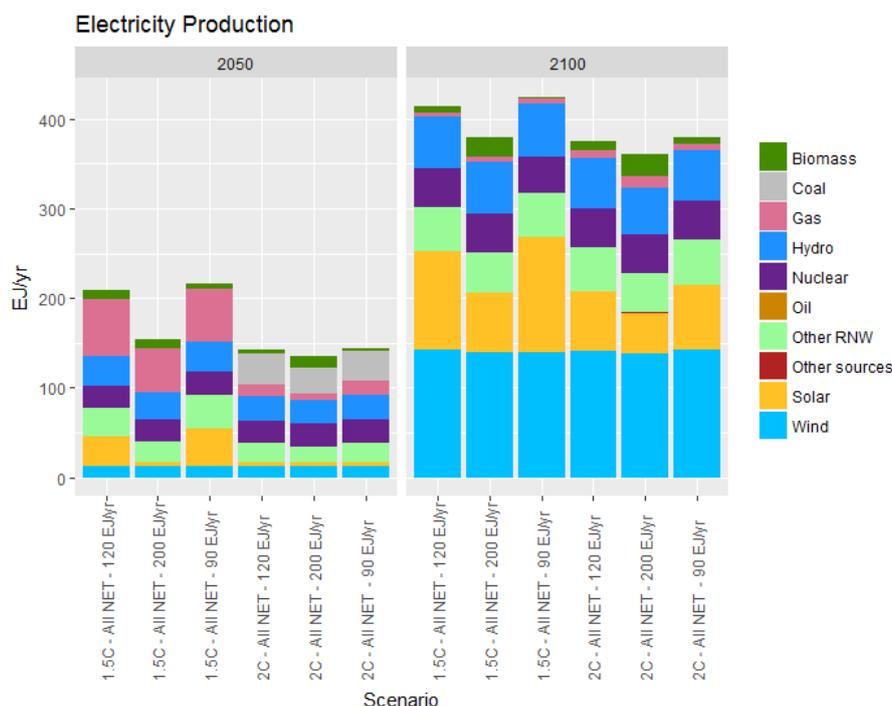
Figure 6.43: Electricity mix for 2°C and 1.5°C scenarios, with a full NET portfolio and different biomass potential.

Considering where bioenergy use is reduced (see table 6.2 below), again it can be seen that the sectors most affected are the electricity and the biofuel production, with massive reduction in 1.5°C scenarios, compared to 2°C target. It is interesting to notice that with a small availability of biomass for 1.5°C, the production of hydrogen from solid biomass is the only sector not being reduced, but even increased of almost 40%.

		COMM	IND	RES	ELC	Biofuel[a]
1st gen Biocrops	1.5C	-	-	-	-55/-80%	-60/ -65%
	2C	-	-	-	-60/-75%	-55/ -60%
2nd gen Biomass	1.5C	-	-	-	0/-25%	-30/ -50%
	2C	-	-	-	-35/-60%	-25/ -40%
End-use Sectors	1.5C	-8/-20%	-15/-25%	-30/-40%	-60% [b]	-
	2C	-10/-20%	-20/-30%	-35/-45%	-50/-60% [c]	-

Table 6.2: Reduction in primary biomass commodities flow to different sectors, when a limited bioenergy potential is applied, for 2°C and 1.5°C with only DAC.a full NET portfolio

[a]Both biodiesel and FT synthesis

[b]These are represented by biofuels for electricity sector)

[c]these are represented by biofuels for electricity sector)

Regional Breakdown

Considering the regional distribution of CDR capacity and the countries most affected by the limited availability of bioenergy (Figure 6.44), it can be seen how in Former Soviet Union the amount of BECCS installed is deeply reduced, as well as in Central South America and USA, while it is not replaced by a similar amount of DAC capacity in the same regions. Indeed, we can notice an increase of Air Capture in Mexico, China and Australia (this is more evident in 1.5°C scenarios).
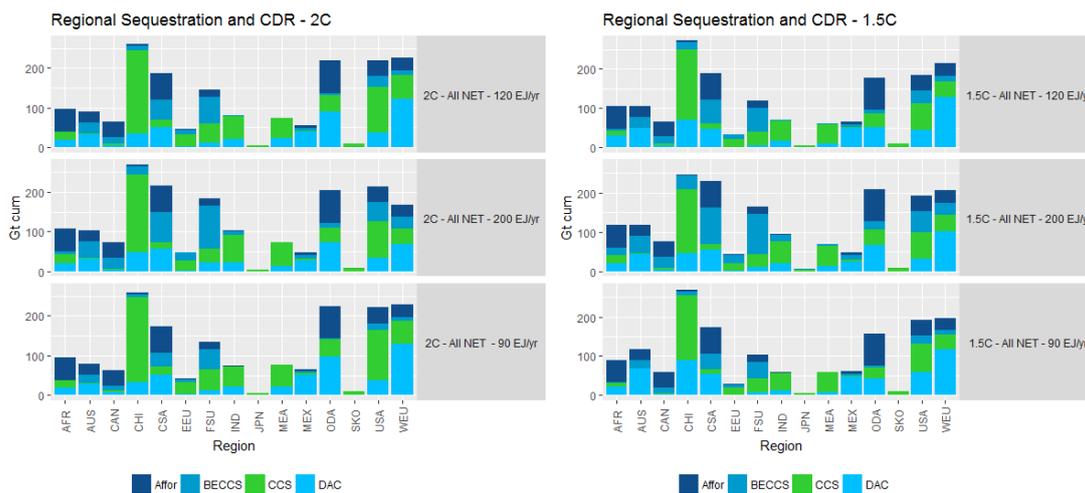
Figure 6.44: Regional breakdown for different sequestration options. Cumulative capture is considered [Gt_{cum}], for 2°C and 1.5°C, with a full NET portfolio and different biomass availability.

Chapter 7

Conclusions and Future Work

Having presented and discussed extensively the results of a wide range of scenarios, in this chapter the main model outcomes and trends will be summarized, considering the initial research questions, with a focus on future work.

7.1 Conclusions

Direct Air Capture Technologies

DAC is still in its early stage so that no technological convergence has been reached yet: different plant designs and sorbent materials are being tested both at lab scale and in first demonstration plants, trying to identify the most suitable one.
Considering also experts opinion through the EE exercise, the designs which are closer to the commercial scale are the one based on hydroxide solutions, currently investigated by *Carbon Engineering* company, and the one adopting amine-modified solid sorbents, as it is being developed by *Climeworks* in Switzerland and *Global Thermostat* in California. All these companies are currently running their pilot plants and they claim to be able to bring DAC on the market by 2025, making it a competitive technology also from a commercial perspective, beside its mitigation role. Their cost estimates are still quite high, around 200-300 \$/ton$_{CO_2}$, but they foresee a significant cost reduction in next years thanks to economies of scale and learning-by-doing mechanisms.
Other materials are being tested to capture CO_2 from the atmosphere, especially solid sorbents, but they have not been deployed at a demonstration scale, so that their real potential is still uncertain and not supported by detailed techno-economic assessment: this is the case for *artificial trees* developed by Lackner. It is likely that new designs will be developed in the future, with further cost reduction potential, as this research field is gaining more and more attention.

The Role of DAC in Stringent Mitigation Pathways

Considering the results discussed in Chapter 5, Negative Emission Technologies appear essential to reach stringent mitigation targets reducing the burden of deep decarbonization for the whole society: indeed, trying to keep the temperature increase

below 2°C and 1.5°C not relying on any CDR option results in extremely high levels of carbon price (i.e. marginal abatement cost), reflecting the infeasibility of these mitigation pathways. Results suggest that an integrated portfolio including Direct Air Capture allows to reduce the mitigation effort in some sectors which are difficult to be decarbonized or with high energy intensity, such as transportation and industrial one, with less drastic decarbonization to be realized in next decades.

In particular it is interesting to underline how a technology which is likely to be deployed only later in the century can have an impact in the short term, mainly looking at the energy mix, thus influencing future investment decision: indeed, integrated models allow to connect long-term temperature targets with policy implementation and strategic investments to be taken in the next decade. According to model results, DAC will become a competitive mitigation option only in the second half of the century, when its capture capacity will reach a Gton scale. Nevertheless, DAC future deployment affects the electricity mix already in 2030, allowing fossil fuel to play a role in the electricity mix up to 2050 and containing the share of intermittent renewable generation, reducing the challenges of a large amount of non-dispatchable generation in term of grid management and storage capacity. Considering a 1.5°C target, the availability of DAC will also decrease the overall electricity demand of more than 25%, due to a lower need for electrification in transport and industrial sectors.

The level of DAC deployed depends mainly by the overall cap applied to CDR options, as the model tends to treat it as a backstop option, thus over-installing it: while BECCS is limited to a capture rate around 10 Gt/yr by bioenergy availability, DAC takes the remaining potential available, as no other external limiting factor are applied. Generally, plants based on solid amine sorbents (DAC2) are deployed earlier in time, while the ones employing hydroxide solutions (DAC1) becomes competitive later in the century, when larger amount of natural gas are available at cheap price to fuel them. Moreover, DAC1 is the technology mostly affected by the presence of other competitive NET options, due to the higher energy requirements.

It is interesting to notice the regional characterization of different DAC technologies, related to the different cost of commodities, with DAC1 generally installed in Western Europe, China and Russian area.

Analyzing the role of DAC within a NET portfolio, it has been demonstrated the advantage of combining different strategies to remove carbon dioxide from the atmosphere, ranging from BECCS to DAC and afforestation, so to reduce the risk of relying on one single approach, as well as the economic impact and the sustainability of meeting stringent targets. I can conclude that there is not a real competition between these BECCS and DAC, but they needs to be developed in parallel.

Impact of Parameter Uncertainties

As in previous modeling exercises [7, 8], *expansion constraints* results binding in determining future DAC diffusion pathways within IAMs, even more than *cost* or *energy parameters*, as this technology is treated as a backstop solution to meet the climate target imposed. This is particularly evident in scenarios consistent with 1.5°C

target, while with less stringent budget a cost reduction favors clearly DAC2 in the last decades, while the main obstacle for the diffusion of DAC1 is represented by energy requirements.

Changing the *time discount rate* and inter-generational preferences, results suggest that DAC technologies will be needed more if mitigation is being delayed in time (higher discounting), while more drastic short-term efforts could reduce the need for massive deployment of this technology and corresponding higher mitigation cost. Nevertheless, even when drastic emission reduction is applied in the first decades, DAC will still play a role in the second half of the century to reduce the impact of mitigation on the energy system.

With a reduced CO_2 *storage availability*, priority is given to DAC plants with respect to other sequestration options, as its role cannot be substituted by other technologies to tackle decentralized emissions, resulting in less BECCS and CCS in both electricity and industrial sectors. Similar trends can be found with both mitigation targets.

Considering a limited *biomass potential* due to competition with land and other sustainability goals, it can be seen that the lower the deployment of BECCS as a carbon removal option, the more important the role assigned to DAC plants to achieve the imposed target, increasing its capacity of about 5 Gt/yr.

To conclude, both thermodynamic and economic considerations suggest that in the long run the cost of large-scale air capture will be comparable to the cost of capturing CO_2 from large fixed sources, making DAC an interesting mitigation option. The commercial use of DAC in niche markets would provide a path for it to be implemented, reducing the initial support needed from governments. On the other hand, regulation would trigger large-scale deployment of DAC as an essential climate change mitigation strategy, by developing national and international policy frameworks for its adoption.

7.2 Future Work

All the results discussed so far confirm the urgency of further research on Direct Air Capture technologies, both from a technical perspective to find the most suitable sorbent materials and optimize plant designs, and from a modeling perspective to assess its role in future mitigation pathways, comparing results from different IAMs to check the robustness of future scenarios for DAC deployment across a wide range of model structure and functional form. The modeling assumptions developed for my research work will be implemented in next months in other Integrated Assessment Models, namely WITCH and IMAGE, before submitting these results in a high-impact journal paper, to inform policy makers based on a solid inter-model comparison exercise. At the same time a more structured Expert Elicitation will be developed, building on my first attempt to realize it.

The outcomes of my research will be presented in November at the Integrated Assessment Modeling Consortium (IAMC) Meeting, as a contribution to the discussion on "Deep Mitigation Pathways".

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Appendices

Appendix A

Questionnaire for Expert Elicitation

Different DAC technologies analyzed
- **Aqueous solution of strong bases** (NaOH, Ca(OH)2, KOH
  Reference: American Physical Society 2011 (Sokolow), Baciocchi et al. (2006),
  Carbon Engineering, Coaway

- **Amine-modified solid adsorbent**
  Reference: Goeppert 2012,
  Global Thermostat, Climeworks

- **Ion-exchange membrane (artificial tree)**
  Lackner, 2009
  Center for negative emissions/Kilimanjaro Energy

A.1. Self-evaluation of expertise on different technologies analyzed

	Not Familiar	Basic Knowledge	Good Knowledge	Expert Knowledge	Among Top Experts
Aqueous solution of strong bases (NaOH, Ca(OH)2, KOH)					
Amine adsorbent					
Ion-exchange membrane (artificial tree)					

Do you foresee other promising technologies?
_____

Who do you think should be definitely included in our elicitation?
_____

A.2. Questionnaire

We are interested in understanding what specific conditions will lead to the development of Direct Air Capture (DAC) technologies and their diffusion thereafter. To achieve commercial scale, they will have to become economically competitive in comparison to other mitigation strategies and negative emission technologies (NET, such as bioenergy with carbon capture - BECCS - or afforestation), with or without considering a price on carbon.

A.2.1 Evaluation of the status of the technology and barriers to commercial success

To evaluate the need of substantial advancement to reach the commercial phase, please insert for each technology a number from 1 to 3:

> 1=Current status is excellent.
> 2=Advances are needed.
> 3=Substantial advances are needed.

Then, according to your expertise, identify the main barriers and specify which stage of the RD&D process is most needed to improve these technologies:

> A-BASIC RD&D, includes the development of new sorbent for CO2 capture; improvement of regeneration processes and their efficiency (e.g., developing catalyst, kiln and furnaces,…); reduction of energy need
> B-ENGINEERING AND APPLIED RD&D, includes improvements in the design of such plants; integration with heat recovery processes or other sources; improved equipments (e.g., improved refractory materials for furnace walls, improve reactor design and fuel processing methods);
> C-DEMONSTRATION, includes construction of a pilot project to test capture on large scale while allowing cost reduction and efficiency increase due to scaling up

	Evaluation (1,2,3)	Specific barriers	Type of RD&D (A,B,C)
Aqueous solution of strong bases (NaOH, Ca(OH)2, KOH)			
Amine-modified solid adsorbent			
Ion-exchange membrane (artificial tree)			

Please, indicate which of the identified barriers could not be overcome with an increase in the level of investment in RD&D:

……

A.2.2. *Evolution of DAC energy consumption*

We are interested in evaluate the energy consumption (both thermal and electrical) of different DAC technologies

Please, define the expected electrical and thermal energy requirement of different DAC technologies in GJ/tonCO2 captured.

To minimize the overconfidence bias, we remind you to reason in the following way:

1. Use a pencil and eraser, rather than pen, so that you may revise your answers as necessary.
2. Think of the highest possible value and the lowest possible value. This is your total estimate range.
3. For each technology, provide the 90th percentile estimate of the characteristics in question.
4. Ask yourself if there are any circumstances that would result in a value higher or lower than the value that you have reported. If so, please revise your estimate.
5. For each technology, provide the 10th percentile estimate of the characteristics in question.
6. Ask yourself if there are any circumstances that would result in a value higher or lower than the value that you have reported. If so, please revise your estimate.
7. Having set your 10th and 90th percentile estimates, please provide your 50th percentile estimate, or best estimate.

THERMAL ENERGY NEED (GJ/tonCO2)

	NaOH Ca(OH)2	Amine adsorbent	Ion-exchange
12 GJ/ton			
11 GJ/ton			
10 GJ/ton			
9 GJ/ton			
8 GJ/ton			
7 GJ/ton			
6 GJ/ton			
5 GJ/ton			
4 GJ/ton			
3 GJ/ton			
2 GJ/ton			
1 GJ/ton			

ELECTRIC ENERGY NEED (GJ/tonCO2)

	NaOH Ca(OH)2	Amine adsorbent	Ion-exchange membrane
3.0 GJ/ton			
2.8 GJ/ton			
2.6 GJ/ton			
2.4 GJ/ton			
2.2 GJ/ton			
2.0 GJ/ton			
1.8 GJ/ton			
1.6 GJ/ton			
1.4 GJ/ton			
1.2 GJ/ton			
1.0 GJ/ton			
0.8 GJ/ton			
0.6 GJ/ton			
0.4 GJ/ton			
0.2 GJ/ton			

Which technology is expected to have the **lowest thermal energy** requirement
- Aqueous solution of strong bases (NaOH, Ca(OH)2, KOH)
- Amine-modified solid adsorbent
- Ion-exchange membrane (artificial tree)

Which technology is expected to have the **lowest electric energy** requirement
- Aqueous solution of strong bases (NaOH, Ca(OH)2, KOH)
- Amine-modified solid adsorbent
- Ion-exchange membrane (artificial tree)

A.2.3. Current estimation of DAC cost

Assessing the cost for DAC technologies is not easy, as the proposed estimations in literature may differ widely, while the assumptions they are based on are not always stated clearly.
Now we are providing you an overview of different cost estimates coming from literature and first pilot plant, and we ask you to **evaluate whether you think they are credible or not.** If you think they are over-/under-estimated, please indicate in the last column the realistic order of magnitude in your opinion (half as much, twice as much, x0.5/x2/x3,...)

Source-year	System description	Cost [$/ton]	Is it credible? (YES/NO)	How much should it be? (x0.5/x2/x3,...)
Keith et al - 2006	aqueous NaOH, causticization with lime, calcination	136 60 (contactor)		
Stolaroff - 2008	aqueous NaOH spray tower	53-96 (capture) 140-250 (overall)		
Lackner - 2009	anionic exchange resin, regenerate with moisture swing	200 30 (long term)		
APS, Socolow - 2011	aqueous NaOH, casuticization with lime, calcination	610		
Holmes, Keith - 2012	aqueous NaOH, casuticization with lime, calcination	60 (capture)		
Mazzotti - 2013	aqueous NaOH, casuticization with lime, calcination - optimized APS design	518 - 568		
Zeman - 2014	aqueous NaOH, casuticization with lime, calcination - optimized APS design	309		
Global Thermostat (company)	Amine-based adsorbent	35 - 50		

A.2.4. *Evolution of DAC cost in different mitigation scenarios*

Now, we are interested in analyzing the evolution of the expected cost of DAC technologies under different mitigation scenarios. The aim is then to assess whether capture of CO2 from the atmosphere will become competitive with respect to other mitigation option (traditional CCS, BECCS, afforestation,...) with or without accounting for a carbon tax. Cost competitiveness does not necessarily imply immediate and extensive diffusion of the technology, as there might exist other barriers to diffusion that we will investigate in the subsequent section. For now, let us concentrate on cost improvements.

Capital and operating costs should be estimated considering installing a plant with a reference scale of **1 MtonCO2/yr.** Operating cost includes **only labor, maintenance and consumables, not the cost of fuel and electricity needed.** No incentive or subsidy should be accounted for.

Please, define the expected capital and operating cost of DAC technologies in 2050 in $/tonCO2 captured, under different mitigation scenarios, considering a plant capacity of 1 MtonCO2/yr

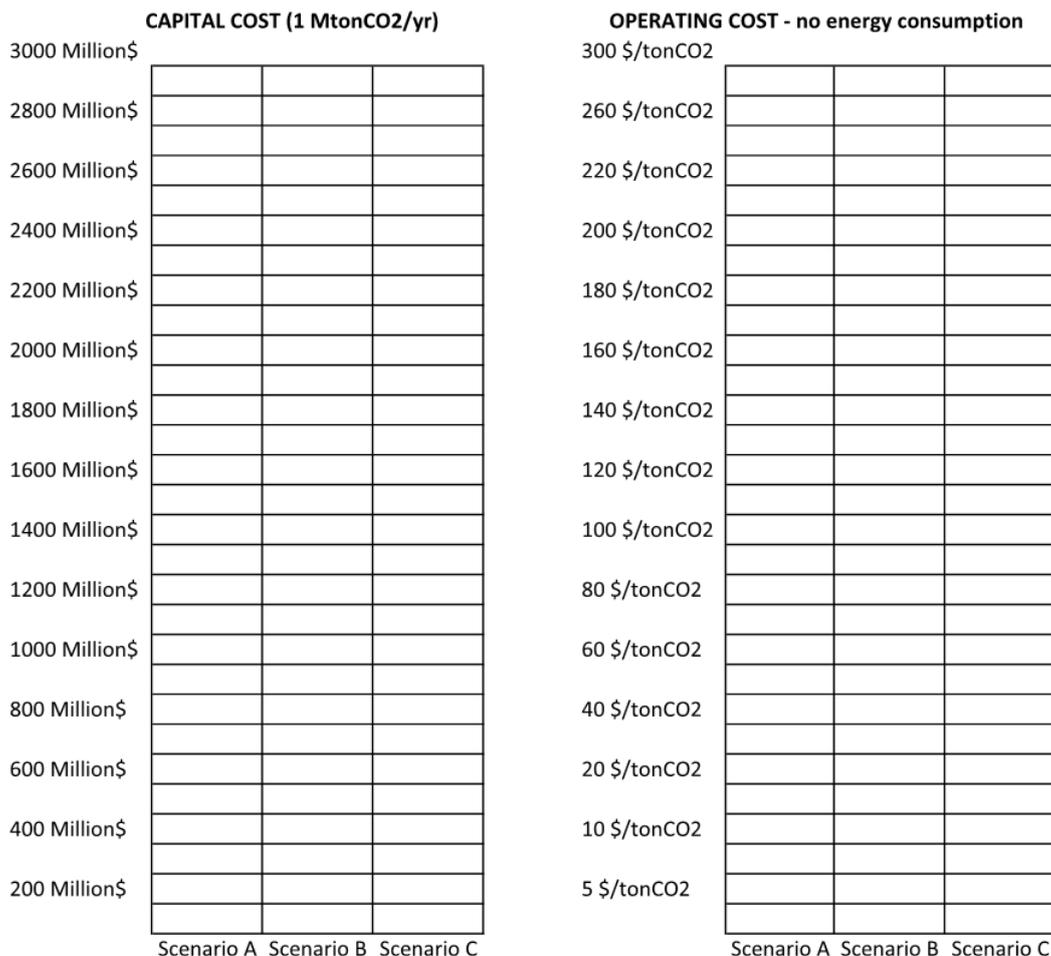To minimize the overconfidence bias, we remind you to reason in the following way:
1. Use a pencil and eraser, rather than pen, so that you may revise your answers as necessary.
2. Think of the highest possible value and the lowest possible value. This is your total estimate range.
3. For each technology, provide the 90th percentile estimate of the characteristics in question.
4. Ask yourself if there are any circumstances that would result in a value higher or lower than the value that you have reported. If so, please revise your estimate.
5. For each technology, provide the 10th percentile estimate of the characteristics in question.
6. Ask yourself if there are any circumstances that would result in a value higher or lower than the value that you have reported. If so, please revise your estimate.
7. Having set your 10th and 90th percentile estimates, please provide your 50th percentile estimate, or best estimate.

Scenario A "no climate policy" baseline (**'business as usual'**). In this scenario, we assume there will be no new global agreement on international climate policy. The energy system will therefore mostly be driven by factors other than climate policy.

Scenario B: Stringent and immediate global climate policy are introduced worldwide in the short term in order to achieve a 50% reduction in global emissions by 2050, with the aim of restricting climate change to a maximum of **2 degrees Celsius.**

Scenario C: Stringent and immediate global climate policy, with the aim of restricting climate change to a maximum of **1.5 degrees Celsius**

COST IN 2050

CAPITAL COST (1 MtonCO2/yr)

	Scenario A	Scenario B	Scenario C
3000 Million$			
2800 Million$			
2600 Million$			
2400 Million$			
2200 Million$			
2000 Million$			
1800 Million$			
1600 Million$			
1400 Million$			
1200 Million$			
1000 Million$			
800 Million$			
600 Million$			
400 Million$			
200 Million$			

OPERATING COST - no energy consumption

	Scenario A	Scenario B	Scenario C
300 $/tonCO2			
260 $/tonCO2			
220 $/tonCO2			
200 $/tonCO2			
180 $/tonCO2			
160 $/tonCO2			
140 $/tonCO2			
120 $/tonCO2			
100 $/tonCO2			
80 $/tonCO2			
60 $/tonCO2			
40 $/tonCO2			
20 $/tonCO2			
10 $/tonCO2			
5 $/tonCO2			

If you had a specific value for the carbon tax when assessing scenario B and C, write it here:

………………………………………………………………………………………………..

Which factor do you believe will affect the most the cost of DAC?
• Solvent choice
• Contactor
• kiln (furnace, oxygen-fired)
• regeneration of the solvent
• O&M costs
• Fuel costs (for thermal energy requirements)
• Power costs (for electric energy requirements)

Which factor do you believe will reduce its cost the most in the future?
• Contactor
• kiln (furnace, oxygen-fired)
• regeneration of the solvent
• O&M costs
• Fuel costs
• Power costs

For each technology, indicate how much it is expected to **reduce its capital and operating cost in 2030 and 2050**, in term of % reduction.

This means, given 100 the cost today, how much this will be in 2030 and 2050 according to you.

CAPITAL COST	Today	2030	2050
Aqueous solution of strong bases (NaOH, Ca(OH)2, KOH)	100		
Amine-modified solid adsorbent	100		
Ion-exchange membrane (artificial tree)	100		

OPERATING COST	Today	2030	2050
Aqueous solution of strong bases (NaOH, Ca(OH)2, KOH)	100		
Amine-modified solid adsorbent	100		
Ion-exchange membrane (artificial tree)	100		

A.2.5. Discussion questions on knowledge spillovers and externalities

Which of the following countries do you think is more likely to be the first to reach a commercially successful breakthrough?
□ Europe
□ USA
□ Japan
□ China
□ Other (specify) ....

Are you concerned about **negative externalities** which might derive from the diffusion of DAC technologies and might impact the environment and society as a whole? Examples of negative externalities might be related to toxic emissions, impact on natural sinks, land and water use,…

………………………………………………………………………………………………

Do you think DAC development could be affected by the **deployment of other technologies** (knowledge spillover)? In positive case, which technology are you thinking at?

………………………………………………………………………………………….

A.2.6. Diffusion

In this last section, we are interested in assessing the conditions that would set back or even prevent the diffusion of DAC technologies, even assuming they have become competitive with respect to other mitigation options.

We have selected a number of factors which could represent the **non-technical barriers** to the diffusion of DAC technologies. Please confirm whether the proposed barriers are important and if necessary please add any further factors of constraint.

Using the table below assess the importance of each of the following factors limiting the diffusion of DAC technologies, by providing a number from 1 (low) to 3 (high). Please also select from the following list the **potential solutions** to overcome the barriers that you consider as the most important and if necessary add specific comments. The suggested solutions include:

- **PI** = policy interventions,
- **AI** = additional investments,
- **ED** = education,
- **MK** = marketing.

Potential barriers	Importance of the barrier 1 Low 2 Medium 3 high	Possible solutions and comments PI policy interventions AI additional investments ED education MK marketing
Long-lived capital (lock-in effect of past investment)		
Rare material supply		
Land availability		
Geographical constraint		
Storage availability		
Public acceptance		
Other: …………		

In this next section, assume that in 2030 DAC technologies will be technically ready to compete with other mitigation option. Considering the non-technical barriers that you have previously identified, we now ask you to provide your **estimates on the diffusion trend** of the bioenergy technologies in power generation.

How many plants with individual capacity of 1 MtonCO2 can be installed each year, once cost-competitiveness has been reached?

………...

Indicate the probability of 3 different diffusion level in term of **annual amount of CO2 captured by DAC at global scale in 2050**, under different mitigation scenarios. Refer to the scenarios presented before:

Scenario A "no climate policy" baseline ('**business as usual**'). In this scenario, we assume there will be no new global agreement on international climate policy. The energy system will therefore

mostly be driven by factors other than climate policy.

Scenario B: Stringent and immediate global climate policy are introduced worldwide in the short term in order to achieve a 50% reduction in global emissions by 2050, with the aim of restricting climate change to a maximum of **2 degrees Celsius.**

Scenario C: Stringent and immediate global climate policy, with the aim of restricting climate change to a maximum of **1.5 degrees Celsius**

With the aim to facilitate the understanding of the proposed numbers, keep in mind that currently the amount of global emission is around 36 GtonCO2/yr and a 500 MW coal power plant emit 1.5 MtonCO2/year We would like these three development levels to loosely represent all possible options, so we ask you to ensure that the sum of probabilities is 100% for each scenario.

	Amount of CO2 captured by DAC at global scale in 2050 [Gton CO2/yr]				
	0 – 5 Gton/yr	5 - 10 Gton/yr	10 - 20 Gton/yr	> 20 Gton/yr	
Scenario A					= 100%
Scenario B (2 °C)					= 100%
Scenario C (1.5°C)					= 100%

Since the date of invention, technologies can experience slow adoption or fast adoption. The figure below portrays a typical s-curve describing technology adoption. If adoption is slow, many years elapse between the date of first market   appearance and the adoption of technology.

(b) Technology diffusion
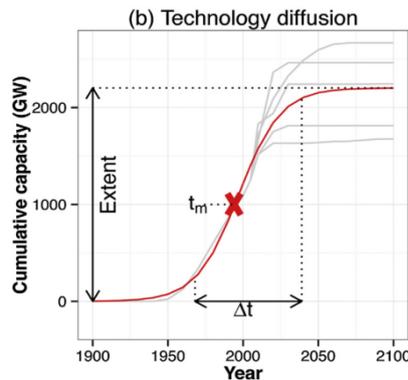
The diffusion trend of DAC technologies will eventually reach a ceiling, as illustrated by the above figure. Can you specify what this **ceiling** may be, in term of cumulative installed capacity in 2100 [Gton CO2 captured/yr] ?
(we are referring to the *Extent* value in the figure)

…………………………………………………………………………………..

What you believe will be the major cause of this ceiling?

…………………………………………………………………………………..

Once technology reaches maturity in 1st country, what do you think is the time (in years) needed to reach the 90% of the ceiling that you indicated before?

…………………………………………………………………………………..

According to the IPCC Fifth Assessment Report (AR5), Negative Emission Technologies (NET) plays a big role in decarbonization in the majority of the scenarios analyzed. In particular, BECCS is being deployed from 2020 on, reaching a rate of removal of 10 GtonCO2/yr in 2020 and 20 GtonCO2/yr. This implies a high development rate expected in next years for all NETs.

What do you think is the closest energy technology to DAC, in term of development pattern (i.e. that experienced a similar diffusion rate in past years)?

…………………………………………………………………………………..

Do you think that in the past there were other technologies that can be compared to DAC expected development rates to meet climate target, also outside the energy sector?

…………………………………………………………………………………..